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Publication Date

2024-03-06

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FISSION OF MEDIUM WEIGHT ELEMENTS

By

Roger Elwood Batzel
B. S. (University of Idaho) 1947

DISSERTATION

Submitted in partial satisfaction of the requirements for the degree of

DOCTOR OF PHILOSOPHY

in

Chemistry

in the

GRADUATE DIVISION

of the

UNIVERSITY OF CALIFORNIA

Approved:

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Committee in charge

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FISSION OF MEDIUM WEIGHT ELEMENTS

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PREFACE

The acceleration of charged particles to energies in the hundreds of Mev range by the Berkeley 184-inch cyclotron has made it possible to study a large variety of nuclear reactions. The characterization of the products from these reactions is done by chemical separations of elemental fractions and subsequent identification of the nuclides formed.

This dissertation includes the results of work on two types of nuclear reactions, namely fission and spallation reactions. In Chapter I evidence is presented which indicates that large fragments (much larger than alpha particles) are emitted among the competitive products of transmutation throughout the entire range of atomic numbers of the elements. Threshold considerations for the observed nuclear reactions show that the reactions are observed with small cross sections well below the threshold for spallation reactions in which the maximum number of alpha particles are considered as being emitted from the excited nucleus. The calculated thresholds include the mass difference between the reactants and the products and the excitation energy which the product particles (or fragments) must have in order to pass over the coulombic barrier. Preliminary experiments on the ranges of recoil fragments from copper irradiated with 340-Mev protons give additional evidence for the emission of heavy fragments. It is suggested that the term "fission" is proper for such reactions throughout the entire range of atomic numbers in which the nucleus is split essentially into pieces of comparable weight.

Chapter II contains the results of the study of nuclear reactions induced in elemental copper by irradiation with 340-Mev protons, and the reaction products cover a range from the region of the target nucleus to a region which is removed from the target nucleus by as many as forty or more nucleons. The yields of the various products were measured, and the results show that a large majority of the products result from reactions in which only a fraction of the total energy of the incident proton is left with the nucleus. The distribution of the amounts of the reaction products is in agreement with a picture of high energy nuclear reactions involving nuclear transparency and the idea that the nuclear reactions involve excitation following collisions and energy transfers between impinging protons and the individual nucleons in the target nucleus.

ACKNOWLEDGMENTS

I wish to thank Professor G. T. Seaborg for his advice and encouragement during the course of this work. I am indebted to Dr. J. G. Conway for the spectrographic analyses of the materials for the irradiations. I also wish to thank Mr. James T. Vale and the group operating the 184-inch cyclotron for the irradiations performed in the course of these studies.

CHAPTER I

NUCLIDES FORMED BY FISSION REACTIONS

I. INTRODUCTION

The fission reaction has been observed with high energy accelerator projectiles for elements as light as tantalum¹ but has not been reported for medium weight elements. Evidence is presented here for occurrence of reactions which are probably most properly described by the term "fission" and which seem to occur with very small yield throughout the entire region where this type of reaction is only slightly exoergic or even endoergic with respect to mass balance.

In the course of the detailed investigation of the spallation of copper and the variation of the product yields with energy of the bombarding particle, the threshold for formation of radioactive Cl^{38} (39-minute half-life) from elemental copper was studied. The energetically most economical way in which Cl^{38} might be formed by spallation reactions is by emission from the bombarded copper nucleus of nucleons in groups such as alpha particles instead of single nucleons. The energetic requirement for the spallation reaction $\text{Cu}^{63}(\text{p}, \text{pn}6\alpha)\text{Cl}^{38}$, in which the maximum number of alpha particles are emitted, is roughly 110 Mev. This threshold includes (1) the mass difference between the reactants and the products, and (2) the excitation energy which the alpha particles must have in order to pass over the coulombic barrier.

The production of Cl^{38} was definitely observed at proton bombardment energies beginning at about 60 to 70 Mev with a cross section of some 10^{-32} cm^2 . The cross section increases rapidly to a value of about 10^{-30} cm^2 at 105 Mev and then increases gradually to a value of about 10^{-27} cm^2 at 340 Mev. In order to explain the low threshold, it must be assumed that substantially

larger particles than alpha particles are emitted from the excited nucleus and the reactions are therefore of a type which might more properly be termed fission. As an example the extreme reaction $\text{Cu}^{63} + p \longrightarrow \text{Cl}^{38} + \text{Al}^{25} + n$, which is energetically most economical but still endoergic, has a threshold of about 50 Mev.

This result made it seem worthwhile to investigate another such reaction in copper and to extend the threshold studies to other elements in the middle portion of the periodic system in order to see whether analogous reactions might occur as a general rule. The variation with energy of the yields of radioactive Na^{24} from copper and radioactive $\text{Sc}^{43,44}$ from bromine were studied. The formation of radioactive Co^{61} from silver, radioactive Na^{24} , Ga^{66} , and Ga^{72} from tin, and radioactive Ga^{66} and Ga^{72} from barium were also studied. In all cases the observed thresholds were well below the thresholds calculated for the reactions in which the maximum number of alpha particles are emitted from the nucleus. Additional evidence for fission of the medium weight elements was found from some preliminary experiments on the ranges of recoil fragments from copper irradiated with 340-Mev protons.

II. PROCEDURE

Irradiations with high energy protons were carried out in the circulating beam of the 184-inch frequency modulated cyclotron, and the proton energy was adjusted by varying the radial distance of the target from the origin of the beam. Foil targets 5 to 10 mils thick were used in the cases of copper, silver, and tin. The bromine was bombarded in the form of ammonium bromide powder wrapped in special purity aluminum foil, and the barium was bombarded in the form of barium carbonate wrapped in special purity aluminum foil. Since the degradation of the energy of the high energy protons is small in traversing the targets used, all targets can be considered thin targets.

The special purity ammonium bromide was synthesized by forming hydrogen bromide from very pure phosphorous tribromide and mixing the gas with very pure gaseous ammonia in a cooled plastic container. The barium carbonate was prepared from special purity barium chloride received from Drs. A. J. Keyes and A. J. King of Syracuse University.

Determinations of the intensity of the internal, proton beam for the silver bombardments and the tin bombardments in which the yields of Ga^{66} and Ga^{72} were studied, were made by bombarding aluminum foils in conjunction with the regular target foils. This monitor foil undergoes a nuclear reaction $\text{Al}^{27}(\text{p},\text{n}3\text{p})\text{Na}^{24}$ for which the cross section has been determined as a function of the energy of the bombarding proton.² The cross sections for the formation of the Na^{24} and the Cl^{38} from copper were determined by comparing the amounts of the isotopes formed with the amounts of radioactive Cu^{64} formed by the $\text{Cu}^{65}(\text{p},\text{pn})\text{Cu}^{64}$ reaction in a given bombardment. The cross section for the reaction $\text{Cu}^{65}(\text{p},\text{pn})\text{Cu}^{64}$ as a function of the energy of the bombarding proton has previously been determined.³ The cross sections for the formation of the $\text{Sc}^{43,44}$ from bromine, the Na^{24} from tin, and the Ga^{66} and Ga^{72} from barium were calculated using an estimated value of 1/2 microampere for the beam current.

After irradiation, the targets were dissolved and the elemental fractions for the various nuclides separated by the chemical procedures to be discussed in detail in the Experimental Details section (V below). Separation procedures were designed to give a radiochemical purification factor of about 10^8 from all other activities formed in the target.

Counting of the activities was done on an end-window, alcohol-quenched, argon-filled, Geiger counter tube with a mica window of $\sim 3 \text{ mg/cm}^2$ thickness used in conjunction with a scale of 64 counting circuit. The nuclides

were characterized by half life determinations, absorption measurements, and sign of particulate radiation emitted.

A crude beta-ray spectrometer was used in the determination of the sign of the beta particles and was especially useful where nuclides with similar half life but differing in sign of particle had to be resolved.

Thresholds for the various reactions include the mass difference between the reactants and products and the excitation energy which the nuclear fragments or alpha particles must have in order to pass over the potential barrier. Where available, measured mass values for the reactants and products were used in making the mass balance. The masses of radioactive fragments formed or considered as being formed as products of the nuclear reaction were calculated by adding the mass equivalent of the decay energy of the radioactive nuclide to the mass of the stable daughter nuclide.

The excitation required for the passage over the potential barrier was calculated assuming that the fragments are spherical and tangent at the nuclear radii (taken as $1.48 \times 10^{-13} A^{1/3}$ cm). Calculations of the coulombic energy requirements for reactions in which a number of alpha particles are emitted were made on the basis that the alpha particles come out consecutively. The coulombic requirement for each alpha particle was calculated on the basis of the alpha particles being tangent to the daughter nucleus and as an example, for the reaction $\text{Cu}^{63}(p, p\alpha)\text{Cl}^{38}$ the first alpha particle is assumed to be tangent to Co^{59} at the nuclear radius, the second tangent to a Mn^{55} nucleus, etc.

The experiments on the determination of the apparent average ranges of the recoil fragments from copper irradiated with 340-Mev protons were

done in the circulating beam of the 184-inch cyclotron. Pieces of nylon (0.5 x 2.5 cm) were placed on both sides of the copper foil (0.5 x 2.5 cm) to catch the recoil fragments. After irradiation, the pieces of copper and nylon were dissolved, carriers added, and the elemental fractions were separated and measured.

The ranges of the recoil fragments were calculated on the basis of an isotropic distribution and constant range for the recoils. The copper foils were thick relative to the range of the fragments observed. The ratio of the number of atoms of a nuclide appearing in the nylon catcher foils to the number of atoms in the copper were related to the ranges by the following treatment of the data.

By integration, the expression,

$$0.5 \int_0^R (R-x/R) dx = 1/4R$$

where R is the range of the fragments in the copper and x is the distance from the surface of the copper, shows that 1/4 of the fragments formed in a thickness R of the surface of the copper will appear in the catcher foils. The relationship $R = (4A_R/A_{TOT})T$, where R is the range of the recoil fragment, A_R is the activity collected in the catcher foil, A_{TOT} is the activity in the copper foil plus that caught on the nylon, and T is the thickness of the copper, gives the range R of the recoils in terms of the information determined experimentally.

III. RESULTS

A. Radioactive Products from Copper

Radioactive Cl³⁸.— The calculated threshold for the spallation reaction $\text{Cu}^{63}(\text{p}, \text{pn}6\alpha)\text{Cl}^{38}$ is about 110 Mev while the threshold for the extreme reaction $\text{Cu}^{63} + \text{p} \longrightarrow \text{Cl}^{38} + \text{Al}^{25} + \text{n}$ is about 50 Mev.

A plot of the cross section for formation of Cl³⁸ from copper is given in Fig. 1. The cross section rises rapidly from 70 Mev to 105 Mev and then increases gradually to a value of $4 \times 10^{-29} \text{ cm}^2$ at 346 Mev. Below 70 Mev the level of activity formed was so low that identification of the Cl³⁸ was not possible, but above 70 Mev the nuclide was identified in all cases through chemical separation, measurement of half-life with a Geiger counter, and observation of the sign of the beta particles with a simple beta-ray spectrometer.

With cross sections so low, an immediate question is that of impurities since a small amount of impurity with a high cross section for formation of Cl³⁸ might be a source of the observed activity. Special purity copper foil was used and a spectrographic analysis submitted with the copper shows less than 0.00007 percent iron, less than 0.0001 percent nickel, less than 0.00005 percent chromium, and less than 0.0001 percent sulfur (the sulfur analyzed chemically). As an additional check, radioactivation methods were also used to check for all possible impurities which might account for the observed yield of Cl³⁸. The results of the activation experiments are listed in Table I.

Except for the value listed for scandium, the cross sections for formation of the Cl³⁸ listed in column two are experimental results from irradiations of the corresponding elements in column one. The cross section value for scandium was estimated from the observed cross sections for

Table I

Examination of Possible Impurity Contribution to Observed Cl³⁸ Activity from Copper

Element	Cross section for formation of Cl ³⁸ at 85 Mev	% Impurity necessary to explain observed activity	% Impurity determined in observed	Ratio of Cl ³⁸ * to Cl ³⁴ formed in bombardment of element
K	<10 ⁻²⁷ cm ²	>0.001	<0.01 (Spect.)	0.06
Ca	<10 ⁻²⁷	>0.001	<0.0001(Spect.)	0.10
Sc	<10 ⁻²⁷	>0.001	<0.0001(Rad.)	
Ti	<10 ⁻²⁸	>0.0016	<0.0001(Rad.)	10.0
V	<10 ⁻²⁸	>0.01	<0.0001(Rad.)	
Cr	<10 ⁻²⁹	>0.1	<0.01 (Spect.)	

calcium and titanium. Column three lists the amounts of the different elemental impurities which must be present in the special purity copper to produce the number of atoms of the Cl³⁸ observed in the chlorine fraction isolated from the irradiated copper. Column four lists the limits set on the actual amounts of the elemental impurities present in the special purity copper as determined by spectrographic and radioactivation methods. Column five gives the ratios of the amounts of the Cl³⁸ to the amounts of Cl³⁴ observed in the chlorine fractions separated from the respective elements after irradiation with 85 Mev-protons. The ratios listed may be compared with the ratio of two observed in the chlorine fraction separated from the special purity copper irradiated with 85-Mev protons.

Potassium as a possible impurity in the copper, which might be a source of the observed Cl³⁸, can be eliminated on the basis of the ratio of the number of atoms of Cl³⁸ formed compared to the number of atoms of Cl³⁴

formed in bombardment of potassium with 85-Mev protons as shown in Table I, aside from the fact that potassium is not a likely impurity in electroplated copper. Calcium impurity was eliminated as a possible source of the activity by considerations shown in the above table: amounts of calcium present, cross section for formation of Cl^{38} , and ratio of Cl^{38} to Cl^{34} . Scandium and titanium were eliminated as possible impurities in the copper by cross section considerations and results of radioactivation analyses performed by irradiating the copper with 30-Mev protons and determining the amount of radioactive Sc^{44} formed. This separation of Sc^{44} also served as a check on the calcium since Sc^{44} could be formed by a (p, xn) reaction on calcium. The radioactivation analysis for vanadium was performed by comparing yields of radioactive Cr^{51} formed in 30-Mev proton bombardments of vanadium with the amount of Cr^{51} formed in the copper foil under similar bombardment conditions. The very small cross section for formation of Cl^{38} as listed in Table I makes it unlikely for chromium or any of the heavier medium weight elements to be present in amounts which would contribute to the Cl^{38} observed since their cross sections for formation of Cl^{38} in bombardments with 85-Mev protons would be lower than that obtained for chromium.

Radioactive Na^{24} . -- The threshold for the reaction $\text{Cu}^{63}(p, p3n9\alpha)\text{Na}^{24}$ should be about 170 Mev of which 100 Mev is due to mass difference and 70 Mev to the coulombic requirement. For the extreme reaction $\text{Cu}^{63} + p \longrightarrow \text{Na}^{24} + \text{K}^{39} + n$, the calculated threshold is about 50 Mev.

The Na^{24} was characterized by absorption and half-life measurements. A plot of the cross section for formation of the Na^{24} against the energy of the bombarding proton is shown in Fig. 1. In addition to the fact that no amounts of impurities were found in the copper which could explain the

activity observed, the fact that the cross section drops so rapidly with decreasing energy of the bombarding proton is also a good indication that the Na^{24} is not formed from an impurity. The cross section determined at 50 Mev is an upper limit since the observed activity could not be characterized definitely.

B. Radioactive Sc^{44} from Bromine

The threshold for the reaction $\text{Br}^{79}(\text{p}, \text{p}7\text{n}7\alpha)\text{Sc}^{44}$ should be about 190 Mev of which ¹²⁰70 Mev is due to mass difference and ⁷⁰120 Mev is required for the potential barrier. For the extreme reaction $\text{Br}^{79} + \text{p} \rightarrow \text{P}^{34} + \text{Sc}^{44} + 2\text{n}$, the calculated threshold is about 80 Mev.

The characteristic 3.9-hour, 1.5-Mev positron disintegration of Sc^{44} was observed in bombardments of bromine with 125- and 140-Mev protons. The chemically separated activity was characterized by absorption and half-life measurements and the observed activity would include any Sc^{43} formed, since resolution of the two 3.9-hour activities was not possible. An attempt was made to find the characteristic activity of Sc^{44} in bromine bombarded with 100-Mev protons, but the small amount of activity could not be identified definitely. In a bombardment at 70 Mev, the scandium fraction showed only 90 disintegrations per minute of an ~ 4-hour activity. Comparable bombardments of bromine with 125- and 140-Mev protons yielded about 10,000 and 30,000 disintegrations per minute, respectively, of the 3.9-hour Sc^{44} . The cross section for formation of the Sc^{44} at 125 Mev is on the order of 10^{-32} cm^2 and is in line with the cross section observed for the formation of Cl^{38} from copper.

The fact that the cross section for formation of Sc^{44} falls off so rapidly as the energy of the bombarding proton is lowered from 140 Mev to 70 Mev rules out the possibility that the observed Sc^{44} is formed from small amounts of calcium, scandium, titanium or vanadium impurities. The

cross sections for formation of Sc^{44} from the elements above vanadium would become increasingly smaller in this energy range so such impurities are not likely. Spectrographic analysis of the ammonium bromide used as target material showed less than 0.001 percent calcium. Scandium, titanium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, zinc, and gallium were not detectable in the spectrographic analysis. The limit set on the amounts of any of these elements present is 0.01 percent. A limit of less than 0.00015 percent iron impurity was set by colorimetric methods.

C. Radioactive Co^{61} from Silver

The production of the nuclide Co^{61} from silver was studied as a function of energy, but the results must be termed borderline due to a small amount of copper impurity present in the silver. Since the Co^{61} is formed as a spallation product of copper, it was necessary to determine the amount of copper impurity present in the silver and the radioactivation method was used. Results of the experiments showed 1×10^{-3} percent copper. The variation with energy of the cross section for formation of the Co^{61} from copper was determined, and the contribution of the Co^{61} formed from the copper impurity was subtracted from the total activity of Co^{61} formed during bombardment of the silver. Resolution of the activity showed that about equal amounts were formed from the silver itself and from the 0.001 percent copper impurity in bombardments at 180 Mev. The observed cross section for formation of Co^{61} from silver at this energy is about 10^{-32} cm^2 .

For the reaction in which the maximum number of alpha particles are emitted, $\text{Ag}^{107}(\text{p}, \text{p}6\text{n}10\alpha)\text{Co}^{61}$, the calculated threshold is about 210 Mev of which about 90 Mev is due to mass difference, and for the reaction $\text{Ag}^{107} + \text{p} \longrightarrow \text{Co}^{61} + \text{Sc}^{45} + 2\text{n}$, the calculated threshold is about 60 Mev.

Attempts were made to continue the cross section work at energies lower than 180 Mev, but the problem of resolving the Co^{61} activity formed from the small amount of copper impurity and that formed from the silver alone becomes increasingly difficult since the cross section for formation of Co^{61} from silver becomes smaller as the energy of the bombarding proton is lowered, while the cross section for formation of Co^{61} from the copper impurity is relatively constant in this energy range.

D. Radioactive Products from Tin

Radioactive Na^{24} .--- The calculated threshold for the reaction in which the maximum number of alpha particles are emitted is about 425 Mev of which 230 Mev is due to mass difference. The extreme reaction ${}_{50}\text{Sn}^{118} + p \longrightarrow \text{Na}^{24} + {}_{40}\text{Zr}^{94} + n$, which is exoergic with respect to mass difference by about 8 Mev, has a calculated threshold of about 50 Mev.

The plot of the cross section for formation of the Na^{24} versus energy is shown in Fig. 2. The cross-section values plotted are good to a factor of two and possibly better since all the irradiations were carried out under conditions where the beam intensity should have been the same.

The point at 75 Mev represents an upper limit for the yield, since the activity in the sodium fraction was too small to identify positively as Na^{24} . The shape of the excitation function almost precludes the possibility of impurities just above sodium in the periodic table. The spectroscopic analysis showed the tin to be 99.999 percent pure with no impurities detectable which could explain the observed activities.

Radioactive Ga^{66} and Ga^{72} .--- The calculated thresholds for the nuclear reactions $\text{Sn}^{118}(p,7n10\alpha)\text{Ga}^{72}$ and $\text{Sn}^{118}(p,13n10\alpha)\text{Ga}^{66}$ are about 230 and 280 Mev, respectively, and for the extreme reactions

$\text{Sn}^{118} + p \longrightarrow \text{Ga}^{72} + \text{Ca}^{45} + 2n$ and $\text{Sn}^{118} + p \longrightarrow \text{Ga}^{66} + \text{Ca}^{49} + 4n$ the thresholds are about 70 and 90 Mev, respectively. The thresholds for formation of Ga^{66} and Ga^{72} from some of the other tin isotopes may be lower by several Mev, but the examples calculated give approximate values upon which to base our reasoning.

The purified gallium fraction isolated from the bombarded tin showed the characteristic activities of Ga^{66} (9.5-hour half-life) and Ga^{72} (14.3-hour half-life), and the absorption curve of Fig. 3 shows the presence of the ~ 3-Mev positron characteristic of Ga^{66} . The gallium activities were identifiable at energies of 150 and 180 Mev with cross sections based on elemental tin of about 10^{-32} cm^2 and with the value at the higher energy a factor of about two times higher than that at the lower energy. Amounts of gallium activity formed in bombardments of tin at 100 Mev were too small to identify definitely and no gallium activity was observed in the gallium fraction separated from tin bombarded with 80-Mev protons.

The spectrographic analysis presented with the special purity tin showed less than 0.0001 percent arsenic, and no zinc, gallium, or germanium were detectable. The variation of the yield of Ga^{66} with energy of the bombarding particle rules out the probability of the observed activity coming from zinc or gallium since the cross section for formation of Ga^{66} from zinc or gallium would rise as the energy is lowered from 180 to 80 Mev, while the yield of Ga^{66} from the tin falls off as the energy is lowered.

E. Radioactive Ga^{66} and Ga^{72} from Barium

The calculated threshold for the reaction $\text{Ba}^{137}(p, 20n13\alpha)\text{Ga}^{66}$ is about 370 Mev (200-Mev mass difference and 170-Mev potential barrier). For the extreme reaction $\text{Ba}^{137} + p \longrightarrow \text{Ga}^{66} + \text{Fe}^{60} + 12n$, the calculated threshold is about 150 Mev. The threshold for formation of Ga^{72} by a

similar extreme reaction is some 40 Mev. lower.

The gallium fraction isolated from barium irradiated with 335-Mev protons showed the characteristic activities of Ga^{66} and Ga^{72} . The cross sections for formation of these nuclides are about equal and are around 10^{-31} cm². The special purity barium contained no spectroscopically detectable impurities with the exception of a trace of iron. Only the single irradiation was performed due to the lack of sufficient special purity barium, but the observed cross section is in line with the others reported and seems to indicate that the gallium isotopes are probably formed by a fission reaction.

F. Evidence from Recoil Experiments

Some recent experiments on the range of recoil fragments from copper bombarded with 340-Mev protons have been done in conjunction with this work. Preliminary results indicate that larger fragments than alpha particles must be emitted from the bombarded nucleus in order to explain the observed energies of the recoils.

Assuming an isotropic distribution for the recoil fragments from the irradiated copper (as discussed in the "Procedure" section (II)), it was found that Cr^{49} nuclei have an average apparent range of about 0.3 mg/cm² of copper against the direction from which the incident protons approach the target, and a range of about 2.5 mg/cm² of copper with the direction of the beam. Using the formulas for stopping power developed by Knipp and Teller⁴ for calculating the energy losses due to electronic and nuclear interactions, the energy of the recoils against the beam was found to be about 1 Mev and with the beam, about 13 Mev.

There is a distortion of the recoil distribution, due to the momentum imparted to the nucleus by the impinging proton, causing more recoil nuclei

to appear with the beam and fewer against the beam. Since the ranges were calculated on the basis of an isotropic distribution for the recoils, the calculated range with the beam is larger and that against the beam is smaller than the actual range. The range distribution was corrected assuming 5 Mev as the kinetic energy that the Cr^{49} fragment received when the excited copper nucleus splits, and assuming that the excited nucleus has had momentum transferred to it by the impinging proton equivalent to 1 Mev of kinetic energy for the excited nucleus. The corrected ranges were about 0.6 mg/cm^2 of copper against the beam and about 2.2 mg/cm^2 of copper with the beam, and are equivalent to energies of about 2.0 Mev and 10 Mev, respectively. These energies correspond to 5.2 Mev of kinetic energy imparted to the Cr^{49} fragment when the excited nucleus breaks up and correspond to a momentum transfer to the copper nucleus equivalent to about 1 Mev of kinetic energy.

A reaction which seems to explain the observed recoil energies is $\text{Cu}^{63} + p \longrightarrow \text{Cr}^{49} + \text{C}^{12} + 3n$. The C^{12} would leave the excited nucleus with a kinetic energy of about 20 Mev which is just equivalent to the height of the potential barrier, and would give a recoil energy to the Cr^{49} of about 5 Mev. Explanation of the observed results in terms of the emission of smaller fragments such as neutrons, protons, or alpha particles would require a great deal of asymmetry in both the energy and distribution of the particles in the process of the break down of the excited nucleus. The observed transfer of momentum to the copper nucleus corresponds to an excitation of the nucleus which would be consistent with the reaction written. In line with the results of the previous sections, it seems reasonable to assume that a reaction of the type written for the formation of Cr^{49} is correct.

IV. DISCUSSION

The coulombic energy requirement for the emission of the charge particles in the above reactions gives rise to a large contribution to the calculated threshold, and the possibility of the alpha particles being emitted at energies well below the top of the coulombic barrier should be considered since in some cases the observed thresholds exceed the values of the mass requirements alone. Calculations of the transmission coefficient⁵ for the coulombic barrier show that the time required for a succession of alpha particles to be emitted with excitation energies well below the top of the barrier is too long to allow this type of nuclear reaction to compete with other reactions in which neutrons and protons (coming out over the barrier) are emitted. It is possible, however, that the coulombic requirement is lowered somewhat due to the effective lowering of the potential barrier at high excitation energies.⁶ The only observed thresholds which are borderline are those of the Co⁶¹ from silver and Ga⁶⁶ and Ga⁷² from barium. The other thresholds are well below the values which could be explained by penetration or an effective lowering of the barrier at the high excitation energies.

The shapes of the excitation curves for the formation of Na²⁴ and Cl³⁸ from copper are quite similar in the low energy region. This similarity could be attributed to the fact that they are formed by reactions which require about the same amount of excitation energy. The extreme fission reactions have about the same threshold and the Na²⁴ fragment corresponds to a very reasonable complementary fragment corresponding to Cl³⁸. The excitation curves for the formation of Na²⁴ and Cl³⁸ from copper probably represent sums of several reactions contributing to the total observed cross section. In the low energy range the main contribution is probably

that of the pure fission reaction while as the energy is raised, reactions in which smaller fragments are given off begin to contribute to the total reaction. At the high energy end spallation reactions probably begin to contribute markedly. The difference in the two curves at the higher energies is probably due to the larger contribution of the direct formation of Cl^{38} by spallation reactions.

It is interesting to note that the emission of nuclear fragments a little larger than alpha particles has been observed in high energy nuclear reactions on medium weight elements. The "hammer tracks" of Li^8 are very well known in reactions induced by cosmic radiation and have also been identified as a reaction product of 340-Mev proton and 190-Mev deuteron bombardments of some of the medium weight elements.⁷ Evidence has also been found for the existence of lithium isotopes as a reaction product of 50- to 340-Mev proton and helium ion bombardments of tin.⁸ Radioactive Be^7 has also been identified as a product of 340-Mev proton bombardments of some of the medium weight elements.⁸ Heavy fragments have also been observed in reactions induced by cosmic radiations.^{9,10,11}

Apparently when the energy threshold requirements are met, large fragments are emitted among the competitive products of nuclear reactions throughout the entire range of atomic numbers of the elements. This is certainly not surprising and the measured yields reported here seem to be quite reasonable. It seems certain that the size of the fragments varies continuously from those (neutrons, protons and alpha particles) which accompany what are conveniently called spallation reactions, through intermediate sizes (for example, Li^8 , etc.), and on up to sizes such that the nucleus is split essentially into several pieces of comparable weight. Apparently a number of reactions in which there occurs the latter type of nuclear splitting have been observed in the present investigation and

perhaps the term "fission" is as proper a name as any to apply to the process. As a result of this work it seems that fission reactions occur in all nuclei if the necessary excitation energy is available.

V. EXPERIMENTAL DETAILS

A. 184-inch Cyclotron Bombardments

All targets were bombarded in the internal beam of the 184-inch cyclotron. Foils were used as target materials in the cases of the copper, silver, and tin; for the bromine bombardments very pure ammonium bromide powder wrapped in special purity aluminum foil was used; and for the barium bombardments, very pure barium carbonate wrapped in the special purity aluminum foil was used. These targets were mounted on the end of a movable probe and the energy of the bombarding proton was selected by an accurate adjustment of the radial distance from the origin of the beam to the leading edge of the target.

The maximum energy of the bombarding proton is a function of the radial distance from the origin of the beam to the leading edge of the target, and since the variation of the energy with radius is known precisely, the precision with which the bombarding energy can be selected depends on the measurement of the distance from the leading edge of the target to the origin of the beam. The measurements are probably good to within 5 Mev at energies of 100 Mev. The spread in energies of the particles for a given radial setting varies from about 3 percent at 348 Mev to 15 to 20 percent at energies around 60 to 70 Mev. This spread in the energy of the particles does not affect the maximum energy of the particles for a given radial setting since the spread is due to the fact that some of the paths of the particles, though circular, have centers which oscillate about the origin of the beam, and the fixed circular path

gives the maximum energy to the particle for a given radial setting of the target.

B. Chemical Separation Procedures

The chemical separation procedures must meet severe requirements since the amount of interfering radioactivities must be reduced to such a degree that the nuclides of interest can be accurately measured by their characteristic radiations. The cross sections for formation of the nuclides produced in the targets bombarded in this series of studies vary from about 10^{-24} cm² down to the observed cross sections of about 10^{-32} cm². Since the number of atoms of the radionuclides which were studied here were extremely small compared to the number of atoms of the interfering radioisotopes, it was necessary that the chemical separation procedures give a radioactive decontamination of at least 10^8 from all other radioactivities formed in the targets.

Sodium from Copper.-- The copper target was dissolved in concentrated hydrochloric acid, hydrogen peroxide was added and 5 mg of sodium chloride was added as carrier. Carriers for the elements zinc through potassium were added, the solution adjusted to 1.5N in hydrochloric acid, and the copper precipitated as the sulfide. The solution was boiled to dryness to remove the excess acid, the residue dissolved, and then the solution made alkaline with ammonium hydroxide. Hydrogen sulfide was added and the sulfides and hydroxides were precipitated. Additional 3 mg portions of carriers zinc through scandium were added and precipitated. This scavenging process was repeated twice. Excess hydrochloric acid was added to the alkaline sulfide solution, the acidified solution boiled to dryness, and the ammonium chloride driven off. The residue was dissolved in water, the solution checked to make sure it was neutral, and the sodium precipitated

as the sodium zinc uranyl acetate. Two 5 ml portions of zinc uranyl acetate were used to wash the precipitate, and the precipitate was then dissolved in absolute ethyl alcohol saturated with hydrogen chloride gas. The resulting solution was cooled in an ice bath and the sodium chloride separated by centrifugation. The sodium chloride precipitate was washed with a 5 ml portion of the alcohol-hydrogen chloride solution. The sodium chloride was dissolved in water and the solution neutralized with potassium hydroxide. The sodium was again precipitated as sodium zinc uranyl acetate, and the precipitate was then washed with additional zinc uranyl acetate and dissolved in absolute alcohol saturated with hydrogen chloride. An additional portion of the alcohol-hydrogen chloride solution was added to wash the precipitate, and then the precipitate was transferred to a plate for counting.

Chlorine from Copper.--- The copper target was dissolved in concentrated nitric acid and 5 mg of chlorine as sodium chloride added to the solution. The chlorine was distilled and trapped in a solution made up of 3 to 4 ml of water containing 2 to 3 drops of concentrated nitric acid, and sufficient silver nitrate to precipitate the 5 mg of chlorine as silver chloride. The resulting solution was heated almost to boiling to coagulate the precipitate. The precipitate was separated and washed with dilute nitric acid. The silver chloride was dissolved in ammonium hydroxide and carriers for zinc, copper, nickel, cobalt, and iron added. The ferric hydroxide precipitate was removed by centrifugation and a slight excess of iodine added as sodium iodide. The silver iodide precipitate was removed, the solution made 0.5N in nitric acid, 5 to 10 mg of bromine added as sodium bromide, the bromide and iodide oxidized with persulfate, and the bromine and iodine

distilled. Silver nitrate was added to the solution and silver chloride was precipitated again. The precipitate was washed with dilute nitric acid and redissolved in dilute ammonium hydroxide. Carriers for zinc, copper, nickel, cobalt, and iron were again added and the ferric hydroxide precipitate removed. The solution was acidified with nitric acid and the silver chloride reprecipitated. The steps for separation of bromine should not be necessary, but were used in order to provide for the eventuality of a very small amount of selenium impurity in the copper which might form some interfering bromine radioactivities during the bombardment of the copper.

Scandium from Bromine.-- The ammonium bromide powder was dissolved in water, 5 mg of scandium carrier added, the solution adjusted to a pH of 3.0, and then transferred to a separatory funnel (consisting of a 40 ml calibrated centrifuge cone with a stopcock sealed to the bottom). Ten ml of 0.5M thenoyltrifluoroacetone in benzene was added and the mixture stirred for 5 minutes to extract the chelate compound which scandium forms with the thenoyltrifluoroacetone. The organic layer containing the scandium was washed 3 times with 10 ml portions of 1N hydrochloric acid. Five mg each of arsenic, selenium, and germanium carriers were added. The solution was made alkaline with ammonium hydroxide and hydrogen sulfide was added to form the polysulfides of selenium, arsenic, and germanium. The scandium precipitates as the hydroxide from this alkaline solution. The precipitate was washed with ammonium sulfide and the scandium dissolved in 6N hydrochloric acid. Iron and gallium carriers were added and extracted from the acid solution with ethyl acetate. Approximately 5 mg of arsenic carrier was added and arsenic sulfide precipitated. Another 5 mg of arsenic carrier was added and the arsenic again precipitated as the sulfide.

The solution was made alkaline with ammonium hydroxide to precipitate the scandium as the hydroxide. The precipitate was dissolved in hydrochloric acid and the solution adjusted to a pH of 3.0. The scandium was extracted with 10 ml of 0.5M thenoyltrifluoroacetone, the organic layer washed with water, and the scandium extracted from the organic layer with 1N hydrochloric acid. The solution containing the scandium was adjusted to 0.5N in hydrochloric acid, and 0.2 ml of 27N hydrofluoric acid was added to precipitate scandium fluoride. The precipitate was washed with 5 ml of 0.5N hydrochloric acid containing 2 drops of 27N hydrofluoric acid. The scandium fluoride precipitate was dissolved in sulfuric acid, the hydrofluoric acid distilled, and the scandium precipitated as the hydroxide.

Cobalt from Silver.-- The silver target was dissolved in nitric acid, 5 mg of cobalt added, and the silver precipitated as silver chloride. Approximately 5 mg of iron carrier was added and the solution made alkaline with ammonium hydroxide. The ferric hydroxide precipitate was washed with hot ammonium chloride and the wash added to the supernatant solution. Approximately 5 mg of strontium carrier was added and strontium carbonate precipitated by adding sodium carbonate. The strontium carbonate precipitation was repeated and the supernatant solution adjusted to 0.3N in hydrochloric acid. Five mg portions of cadmium, palladium, and copper carrier were added to the solution and the sulfides precipitated with hydrogen sulfide. The supernatant solution was made alkaline with ammonium hydroxide and the cobalt precipitated as cobalt sulfide. The precipitate was washed with water to remove the ammonium sulfide and then dissolved in nitric acid. Silver carrier was added to the solution and silver chloride precipitated to remove any silver which might not have been removed. The

solution was made alkaline with potassium hydroxide and the cobalt hydroxide precipitate removed. The cobalt hydroxide was dissolved in acetic acid and potassium cobaltinitrite precipitated by adding an equal volume of a hot saturated solution of potassium nitrite. The precipitate was washed and then dissolved in concentrated hydrochloric acid. The solution was adjusted to 0.1N in hydrochloric acid, and 3 g of sodium thiocyanate added. The thiocyanate complex ion of cobalt was extracted into a one to one mixture of amyl alcohol and diethyl ether. The cobalt was extracted from the alcohol-ether layer with 6N aqueous ammonium hydroxide and the cobalt precipitated as the sulfide. The sulfide precipitate was dissolved in concentrated nitric acid, the solution neutralized with potassium hydroxide and then acidified with acetic acid. The cobalt was precipitated in the final form as potassium cobaltinitrite.

Sodium from Tin.--- The separation of the sodium from the tin is almost the same as that for the separation from copper. Different carriers, ruthenium, palladium, molybdenum, silver, strontium, rubidium, lanthanum, iron, and calcium were added and then precipitated several times to remove the interfering activities. The subsequent sodium precipitations were carried out in the manner given in the section on "Sodium from Copper"

Gallium from Tin.--- The tin target was dissolved in concentrated hydrochloric acid with just sufficient concentrated nitric acid added dropwise to speed the dissolving of the tin. Five mg of gallium carrier and "hold-back" carriers for iron, antimony, ruthenium, and molybdenum were added to the solution. The solution was adjusted to 7.75N in hydrochloric acid and the gallium extracted with isopropyl ether. The ether layer was washed six

times with 10 ml portions of 7.75N hydrochloric acid. The gallium was extracted from the ether layer with water. The water layer was adjusted to 1N in hydrochloric acid and the sulfides precipitated with hydrogen sulfide. Milligram amounts of molybdenum, antimony, and palladium were added to the supernatant solution and the sulfides precipitated. Molybdenum, antimony, and palladium carriers were again added and the sulfides precipitated. Three mg of antimony carrier was added to the supernatant solution and antimony sulfide precipitated. The antimony sulfide precipitation was repeated three more times. The supernatant solution was boiled to remove the hydrogen sulfide and then adjusted to 1N in sodium hydroxide. The ferric ion which follows the gallium through the chemical separations precipitates here as the hydroxide. Three mg of strontium was added and the strontium precipitated as the carbonate. Five mg of iron carrier was added and the hydroxide precipitated. The solution was adjusted to 7.75N in hydrochloric acid and the gallium extracted with isopropyl ether. The ether layer was washed three times with 10 ml portions of 7.75N hydrochloric acid, and the gallium was extracted from the ether layer with water. Five mg of ruthenium carrier and 5 ml of concentrated perchloric acid were added and the resulting solution evaporated to dryness. Five ml of concentrated perchloric acid was again added and the solution again evaporated to dryness. The residue was taken up in 1N hydrochloric acid, 2 mg of antimony carrier added, and the antimony precipitated as the sulfide. Two mg of antimony was again added to the solution and the sulfide precipitated. The solution was boiled to remove the hydrogen sulfide and adjusted to a pH of 5.5 with sodium hydroxide to precipitate gallium hydroxide. The hydroxide precipitate was dissolved and adjusted to 7.75N with hydrochloric acid. The gallium was extracted with isopropyl ether and the ether layer washed

twice with 10 ml portions of 7.75N hydrochloric acid. The gallium was extracted from the organic layer with water and the water layer adjusted to a pH of 5.5 with sodium hydroxide to precipitate the gallium hydroxide.

Gallium from Barium.-- The procedure used for the chemical separation of gallium from the irradiated barium was essentially the same as that described in the section "Sodium from Tin."

Chromium from Copper.-- The irradiated copper or the nylon catcher foils were dissolved in concentrated nitric acid and the nitrate converted to the chloride by boiling with hydrochloric acid. Carriers for the elements calcium through zinc were added, the solution adjusted to 2N in hydrochloric acid, and the copper precipitated as the sulfide. The supernatant solution was made alkaline with ammonium hydroxide and the alkaline sulfides precipitated. The precipitate was dissolved in concentrated nitric acid and manganese dioxide precipitated by adding potassium chlorate. The supernatant solution was adjusted to about 0.1N in nitric acid, and cooled in an ice bath. Hydrogen peroxide was added to form the blue peroxychromic acid which was extracted into diethyl ether. The ether layer was washed four times with 5 ml portions of water acidified with two drops of concentrated nitric acid. Five ml of water made alkaline with sodium hydroxide was added to extract the chromium from the ether layer. The ether was removed by evaporation and the chromium oxidized completely with hydrogen peroxide in the alkaline solution. After boiling to remove the excess peroxide, the solution was made slightly acid and the chromium precipitated in the final form as barium chromate.

CHAPTER II

NUCLIDES FORMED BY SPALLATION REACTIONS

I. INTRODUCTION

The acceleration of charged particles to energies in the hundreds of Mev range by the Berkeley 184-inch cyclotron has made it possible to study nuclear reactions in which the reaction products cover a range from the region of the target nucleus to a region which is removed from the target nucleus by as many as forty or more nucleons.¹² The term "spallation" has been suggested to describe this type of transformation in which the excitation of the nucleus is degraded by emission of small nuclear fragments such as neutrons, protons, deuterons, and alpha particles.

Some of the results of spallation reactions induced with high energy deuterons and alpha particles have been reported previously.¹²⁻¹⁶ The present study consists of the determination of the radioactive products formed by the irradiation of elemental copper with 340-Mev protons.

The observed spallation products include some 35 nuclides from sodium through zinc and the distribution of the amounts of the reaction products formed is in general agreement with the picture of high energy nuclear reactions described by Serber.¹⁷ The observed yields show the effects of nuclear transparency and are consistent with Serber's idea that the nuclear reactions involve excitation following collisions and energy transfers between the impinging proton or the individual nucleons in the impinging particle in the case of deuterons and helium ions and the individual nucleons in the target nucleus. As would be expected on this picture, the products which are formed by reactions requiring small amounts of excitation occur most frequently.

II. PROCEDURE

Irradiations with the high energy particles were carried out in the circulating beam of the 184-inch frequency modulated cyclotron. The targets consisted of 10 mil copper foil about 15 mm long and 10 mm wide. The copper strips were clamped in target holders which could be attached to the movable probe head of the cyclotron. Since the degradation of the energy of the high energy particles in passing through the 10 mil copper foil is only on the order of 0.5 Mev the targets could be considered as thin targets.

Because of the fluctuations in intensity of the circulating beam, the exact amount of the irradiation of the copper target could not be determined directly. The amount of the nuclide Cu^{61} formed during any given irradiation of the copper was determined and by relating the amounts of the other nuclides formed to the amount of Cu^{61} produced, relative yields of nuclides from any series of irradiations could be compared.

The length of the irradiations varied from ten minutes to one hour depending on the half-lives of the nuclides which were to be studied. One approximately 30-hour proton bombardment was done to determine the yields of the nuclides with very long half-lives.

An absolute cross section for the formation of the Cu^{61} from copper irradiated with 340-Mev protons was determined by irradiating a piece of 10 mil copper foil in the external beam of the 184-inch cyclotron. The beam intensity was measured directly by means of a Faraday Cup arrangement.

After the target was irradiated, it was dissolved and measured amounts of carriers were added to the target solution. The elemental fractions were

separated by the chemical separation procedures to be discussed in the section on "Chemical Separation Procedures." The amount of carrier remaining after the chemical separation of an elemental fraction was determined by chemical analysis and the percent of the carrier recovered was taken as a measure of the percent of the radioactive atoms of the element recovered.

Counting of the activities was done on an end-window, alcohol-quenched, argon-filled Geiger counter tube with a mica window of ~ 3 mg/cm² thickness used in conjunction with a scale of 64 counting circuit. The nuclides were characterized by half-life determinations, absorption measurements, and sign of particulate radiation emitted.

A crude beta-ray spectrometer was used in the determination of the sign of the beta particles and was especially useful where it was necessary to resolve nuclides with similar half-life but differing in sign of particulate radiation emitted.

The counting data were corrected for window and air absorption, and in the cases where the yields are based on the counting of K x-rays corrections were made for the fluorescence yield of the x-rays. A backscattering correction was made only in the case of the determination of the absolute cross section for the formation of the Cu⁶¹ since all samples were mounted on cover glasses or 5 mil aluminum and for such backing the corrections are almost constant in the energy range of the beta particles observed. No correction was applied for self-absorption since the weights of the samples counted were almost negligible in all instances.

III. RESULTS

A. Radioactive Nuclides Identified

The characteristic activities of the nuclides discussed in this section were observed in the elemental fractions separated from the irradiated copper target. References for most of the nuclides characterized may be found in the 1948 edition of "Table of Isotopes" by Seaborg and Perlman.¹⁸ Only in the cases where information on the nuclides was published in the literature subsequent to the data given in "Table of Isotopes" will references be cited.

Zinc Fraction.-- The characteristic activities of Zn^{65} , Zn^{63} , and Zn^{62} were observed in the zinc fraction. The Zn^{65} was counted through sufficient aluminum absorber to cut out completely the particulate radiation and x-rays, and the counting efficiency of the 1.1-Mev gamma-ray was taken as 1 percent. The Zn^{63} was characterized by its half-life and the nuclide was assumed to decay 93 percent by positron emission and 7 percent by K-electron capture. The Zn^{62} was assumed to decay 10 percent by positron emission and 90 percent by K-capture¹⁹ and was counted with the 10-minute Cu^{62} daughter in equilibrium.

Copper Fraction.-- The activities resolved in the copper fractions were those of Cu^{64} , Cu^{62} , and Cu^{61} . The 12.8-hour Cu^{64} was the longest lived activity identified and was assumed to have a counting efficiency of 50 percent. The 10.5-minute Cu^{62} was the shortest lived activity identified and the nuclide was considered as decaying completely by positron emission. Cu^{61} decays 66 percent by positron emission²⁰ and 66 percent was taken as the counting efficiency of the nuclide.

Nickel Fraction.--- The only observable activities in the nickel fraction were the 2.6-hour Ni^{65} and the 36-hour Ni^{57} . The formation of the small amount of Ni^{65} from copper irradiated with protons is probably due to the reaction $\text{Cu}^{65}(\text{n,p})\text{Ni}^{65}$ caused by secondary neutrons and will be neglected as a spallation product. A counting efficiency of 50 percent was used for the Ni^{65} since the nuclide decays 50 percent by positron emission and 50 percent by orbital electron capture.²¹

Cobalt Fraction.--- The radioactivities found in the cobalt fraction were identified as Co^{61} , Co^{58} , Co^{56} , and Co^{55} . The Co^{61} decayed with the characteristic 1.75-hour half-life and since it decays by negatron (negative beta particle) emission no correction was necessary for the counting efficiency. The Co^{55} was easily resolvable and since no K-electron capture branching has been reported the counting efficiency was taken as 100 percent.

The fact that the Co^{58} and Co^{56} decay with equal half-lives and are both positron emitters makes their resolution difficult. The shape of the composite positron spectrum from the cobalt fraction, was determined by means of a crude beta ray spectrometer after the 18-hour Co^{55} had decayed. The shape of the positron spectrum for a sample of pure Co^{56} which had been determined under similar conditions was available, and by subtracting the contribution of the Co^{56} positron spectrum from the composite positron spectrum the contribution of each nuclide to the total activity was obtained. The counting efficiency of the Co^{58} was taken as 15 percent and that of the Co^{56} as 100 percent.

Iron Fraction.--- The characteristic activities of the radionuclides Fe^{59} , Fe^{55} , Fe^{53} , and Fe^{52} were identified in the iron fraction separated from the irradiated copper. The 2.9-year Fe^{55} was assumed to decay completely by orbital electron capture²² and a counting efficiency of 10 percent was

taken for the x-rays. The Fe^{53} was assumed to decay 100 percent by positron emission. The Fe^{52} was counted in equilibrium with its 21-minute Mn^{52} daughter. By determining the amount of 21-minute Mn^{52} in equilibrium with the Fe^{52} and milking the 5.8-day Mn^{52} which grows into the iron fraction an estimation was made of the amount of positron and orbital electron capture branching for the Fe^{52} . It was found that the nuclide decays approximately 65 percent by positron emission and 35 percent by orbital electron capture.

Manganese Fraction.--- The radionuclides Mn^{56} , Mn^{54} , Mn^{52} , and Mn^{51} were identified in the manganese fraction. The Mn^{56} was assumed to decay completely by negatron emission. The Mn^{54} was counted through sufficient aluminum absorber to cut out all the particulate radiation and x-rays, and a value of 0.8 percent was assumed as the counting efficiency of the 0.8-Mev gamma-ray. Thirty-five percent was taken as the positron branching of the 5.8-day Mn^{52} , and the yield for manganese mass number 52 is reported on the basis of this activity. The Mn^{51} was assumed to decay completely by positron emission.

Chromium Fraction.--- The two activities identified in the chromium fraction were those of Cr^{51} and Cr^{49} . Since the Cr^{51} decays by orbital electron capture and gamma-ray emission the counting efficiency of this nuclide would have had to be estimated roughly, but it was possible to determine the counting efficiency directly by milking Cr^{51} from a known amount of Mn^{51} which had decayed. With the assumption that Mn^{51} decays 100 percent by positron emission, the Cr^{51} was found to have a counting efficiency of 2.6 percent when counted in a precipitate of barium chromate. The Cr^{49} was assumed to decay with no orbital electron capture branching.

Vanadium Fraction.--- The yield for vanadium is based on V^{48} which was assumed to decay 58 percent by positron emission.

Titanium Fraction.--- The only activity identified in titanium was that of the 3.08-hour Ti^{45} and the yield was calculated on the basis of the nuclide decaying entirely by positron emission.

Scandium Fraction.--- Several activities were observed in the scandium fraction, and these activities were identified as belonging to Sc^{48} , Sc^{47} , Sc^{46} , Sc^{44m} , and a mixture of the 3.9-hour Sc^{44} and Sc^{43} . The 3.9-hour and 85-day activities were easily resolvable from the decay curves, but the resolution of the 2.44-day Sc^{44m} , the 3.4-day Sc^{47} and 44-hour Sc^{48} was more difficult. The contribution of the 2.44-day Sc^{44} , with its 3.92-hour positron daughter in equilibrium, to the total activity was resolved approximately with the crude beta-ray spectrometer. With the positron activity resolved, the remaining activity was almost entirely that of the 3.4-day Sc^{47} , and the small amount of Sc^{48} activity resolved was so uncertain that the yield of this nuclide is not reported. The yield for Sc^{43} includes the 3.9-hour Sc^{44} and these nuclides were assumed to decay completely by positron emission. The yield for Sc^{44} was calculated on the basis of the 2.44-day isomer.

Calcium Fraction.--- Only two activities were observed in the calcium fraction. One was the 150-day Ca^{45} , and the other was a 4.8 ± 0.2 day beta emitter with energy of about 1.2 Mev as determined by an aluminum absorption measurement. This activity is probably the 5.8-day scandium activity reported as Ca^{47} by Matthews and Pool.²³ The growth of a 3.4-day scandium was observed in the decay of the calcium fraction, and the scandium daughter was milked from the fraction. The aluminum absorption

measurement of this 3.4-day scandium daughter showed it to be the activity assigned to Sc^{47} . The decay curve of the calcium fraction is shown in Fig. 4, and Fig. 5 shows the decay of the calcium fraction counted through sufficient aluminum absorber to cut out the beta particles of the Ca^{45} and the Sc^{47} daughter.

Chlorine Fraction.--- The characteristic activities of Cl^{39} , Cl^{38} , and Cl^{34} were observed in the chlorine fraction. The resolution of the Cl^{38} and Cl^{34} was done by determining the relative amounts of positron and negatron present by means of the crude beta ray spectrometer. The Cl^{39} was resolved directly from the gross decay curve. The Cl^{39} , Cl^{38} , and Cl^{34} were assumed to have counting efficiencies of 100 percent.

Phosphorous Fraction.--- The only activity observed in the phosphorous fraction was that of the 14.3-day P^{32} .

Sodium Fraction.--- Two activities were observed in the sodium fraction. The 14.8-hour Na^{24} was definitely identified, but the 2.6-year Na^{22} could be identified only on the basis of the decay during a 120-day period.

B. Yields of Spallation Products of Copper

A plot of the observed yields of the spallation products is shown in Fig. 6. The yields are given relative to Cu^{61} which was arbitrarily assigned a yield of 1.0. The numbers listed from the various nuclides represent the ratios of the number of atoms of the particular nuclides to the number of atoms of the Cu^{61} formed in copper irradiated with 340-Mev protons, and hence the numbers represent the ratios of the cross sections for formation from elemental copper. The cross section for the formation of Cu^{61} from elemental copper bombarded with 340-Mev protons is $1.7 \times 10^{-26} \text{ cm}^2$.

as determined by a bombardment of copper in the external proton beam where the beam intensity was accurately measured. Many of the values for the reported relative yields are very dependent upon the counting efficiencies assumed (e.g., orbital electron capture), and when these efficiencies are better known the yields can be recalculated.

IV. DISCUSSION

A. General Observations

The wide distribution and large number of radionuclides formed as spallation products of copper are immediately apparent from a study of Fig. 6. The identification of Cu^{64} , Co^{58} , Mn^{54} , and Sc^{46} shielded from formation by decay, is good evidence for believing that the observed nuclides are mainly primary products from the splitting up of the excited nucleus. The radionuclides with the largest neutron deficiencies or neutron excess are formed in lowest yield, and the yield for a given Z rises for the nuclides nearest the region of stability. Extrapolation of this effect to the region of stability indicates that the stable nuclides are formed in high yield. Thus the data indicate that the emission of almost exclusively neutrons or protons from an excited nucleus is not probable.

The majority of the observed yield is found in the region of the target nucleus indicating that reactions requiring much less than the maximum amount of excitation available to the nucleus are more probable, and this is discussed in some detail in the next section.

It should be possible, knowing the general distribution of spallation products for a given Z , to extrapolate and interpolate yields for the nuclides not directly observed as spallation products of copper. The data have been treated in this manner as shown in Fig. 7. It was assumed that the most probable yield for a given Z is a region 2 mass units wide and that the

yields 1 mass unit on either side of this region are formed in yields about 1.5 times lower than the values in the region of the maximum yield, that spallation yields 2 mass units removed are about 10 times lower than the maximum, and that yields 3 mass units removed are about a factor of 20 lower than the maximum.

A summation of these extrapolated and interpolated yields for 340-Mev protons gives a total yield relative to Cu^{61} of about 30, and using the measured value of $1.7 \times 10^{-26} \text{ cm}^2$ for the cross section for formation of the Cu^{61} , the total spallation cross section for copper is about $0.5 \times 10^{-24} \text{ cm}^2$. The geometrical cross section for copper is about $1.1 \times 10^{-24} \text{ cm}^2$. The discrepancy corresponds in part to the nuclear transparency at these high energies, but probably to a larger extent it is explained by the approximate nature of this method of estimation of the reaction cross section. The data in Fig. 6 show that about 80 percent of the spallation yields are concentrated in the elements copper, nickel, and cobalt. Outside the immediate region of the nucleus, the yield values in the region of most probable yield for a given Z are a decreasing function of Z indicating that nuclear reactions requiring very high excitations of the nucleus are much less probable than reactions requiring perhaps some 25 to 50 Mev of excitation.

B. Mechanisms of High Energy Spallation

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 compound nucleus with an excitation energy equal to the kinetic energy plus the binding energy of the incident particle. The excitation is then dissipated as a separate step by the evaporation of nucleons to

form the product nucleus. Using only this picture of the compound nucleus, one would expect that the bombardment with particles of several hundred Mev of energy would lead to very low yields of nuclides which are within a few mass units of the target nucleus, since emission of a large number of particles from the highly excited compound nucleus would be much more probable. On the contrary, however, the data presented in the preceding sections show that a large majority of the reactions induced in copper irradiated with 340-Mev protons, lead to products which differ from the target nucleus by a loss of only several nucleons.

Serber has suggested a mechanism which very satisfactorily explains the observed facts.¹⁷ He points out that the collision time between a high energy incident nucleon and a nucleon in the nucleus is short compared to the time of collision of the nucleons in the nucleus, suggesting that collisions between incident nucleons and the individual nucleons in the nucleus are of primary importance.

Since the individual nucleon-nucleon collisions are important, the high energy nuclear reactions can be interpreted in terms of the high energy scattering between free nucleons. Consideration of high energy scattering leads to two conclusions. First, at sufficiently high energies the nucleus becomes partially transparent to the bombarding particles, and second, the incident particle loses only a fraction of its energy in the collisions. Since the struck particles have a shorter mean free path than the incident one, they will usually distribute their energy to other nucleons through collisions, but it is possible that these struck nucleons can escape from the nucleus with little or no energy loss. The subsequent behavior of the excited nucleus can be described in terms of an evaporation model, with the excitation energy dissipated by boiling off particles with several

Mev of kinetic energy each.

The high spallation yields in the immediate region of the target nucleus are probably formed by reactions in which only single nucleon-nucleon collisions take place. Minimum energy transfers from a single nucleon-nucleon collision would take place when such a collision occurs near the edge of the nucleus, and the struck nucleon escapes from the nucleus with little or no energy transfer to the nucleus.

A larger portion of the energy of the incident particle may be left with the nucleus if multiple nucleon-nucleon collisions take place. This would lead in the extreme to products resulting from nuclei excited to almost the full energy of the impinging proton, amounting essentially to the formation of a compound nucleus in the ordinary sense. Thus the target may receive excitation energy from about zero up to the full energy of the projectile.

It is difficult to estimate the energetic requirements for formation of spallation products such as Cl^{38} and Na^{24} . The threshold for formation of Cl^{38} from copper by the reaction $\text{Cu}^{63}(\text{p}, \text{pn}6\alpha)\text{Cl}^{38}$ in which the maximum number of alpha particles emitted is roughly 110 Mev, but recent experimental results also indicate that fragments larger than alpha particles are emitted among the competitive products of nuclear reactions,²⁴⁻²⁶ and the energetic requirements for these reactions are even lower than for alpha particle emission. The observed yield for any given spallation product of copper probably actually represents the sums of the yields of several types of nuclear reactions which form the given nuclide. It is possible, however, in the light of some earlier work to draw some conclusions about the energetic requirements. The observed yields for Cl^{38} from copper irradiated with 190-Mev deuterons and 190-Mev helium ions were found to be about a factor of six lower than those from copper irradiated with 380-Me. helium ions.²⁷

The yield of Cl^{38} from the copper irradiated with 340-Mev protons is comparable to that from the copper irradiated with 380-Mev helium ions. These differences in yields indicate that nuclides in the region of Cl^{38} are probably formed mainly by reactions which require an appreciable part of the entire energy of the incident high energy particle.

It is not difficult to visualize, in terms of an average energy loss of about 25 Mev per nuclear collision and a mean free path ($4 \times 10^{-13} \text{ cm}^2$) of about $2/3$ the nuclear radius for copper,¹⁷ mechanisms by which 190-Mev deuterons and 190-Mev and 380-Mev alpha particles might impart large portions of their total energy to the copper nucleus. It is difficult, however, to understand how a 340-Mev proton could impart large amounts of excitation to the nucleus with a fair probability if it transfers in a single nucleon-nucleon collision an average kinetic energy only slightly higher than the value of 25 Mev given by Serber for 100 Mev nucleons and has a mean free path about equal to the nuclear diameter for copper as estimated by Yamaguchi.²⁸

In the light of some recent results of high energy scattering experiments,^{29,30} it is possible to actually estimate the mean free path of 340-Mev protons in nuclear matter and to estimate the average kinetic energy loss of a 340-Mev proton in one individual collision with a nucleon in a copper nucleus and to see if the values estimated for the average kinetic energy loss and mean free path are more in line with high energy transfers having a fair probability.

The experimental data on proton-proton scattering at 340 Mev²⁹ show that the scattering cross section does not follow an expected $1/E$ dependence, and that the cross section is considerably higher than would be expected if it varied as $1/E$. Also the results indicate that the cross section is practically isotropic between 0° and 90° in the laboratory system. This

means that high energy transfers in single proton-proton collisions have a relatively high probability. By extrapolating the data from neutron-proton scattering with 260-Mev neutrons³⁰ to 340 Mev and averaging the cross sections for N-P and P-P scattering at the different scattering angles, it is possible to estimate a mean energy loss for the 340-Mev protons of about 70 Mev per collision in a copper nucleus.

On the basis of these scattering data the mean free path of a 340-Mev proton in nuclear matter has been estimated to be about the same as that for a 95-Mev nucleon, about 4×10^{-13} cm.³¹ On the basis of the mean free path of the 340-Mev protons being about 2/3 the length of the nuclear radius for copper, and on the basis of the mean kinetic energy transfer per single nucleon-nucleon collision being about 70 Mev, it is not difficult to understand how large amounts of excitation are imparted to the nucleus.

Recent calculations based on an evaporation model for the emission of the particles from the excited nucleus indicate that the emission of alpha particles from the excited nucleus should be a fairly probable occurrence and that the evaporation of particles should lead to products along the region of stability.^{6,32} The observed spallation yields are in agreement with these calculations since the yields apparently do have a definite maximum of most probable yield for a given Z about 2 mass units wide and a line drawn along this region of most probable yield would be drawn through the region of stability. The very high yield of Mn⁵² compared to Fe⁵² for the isobaric pair Fe⁵²-Mn⁵² indicates that there is a much larger difference in the yields than would be expected from the regular trends of the spallation yields, and it seems logical to explain the difference observed in terms of alpha particles being boiled off from the excited target nucleus as suggested by Miller²⁷ and Helmholtz et al.¹⁶ If

the Fe^{52} and Mn^{52} are formed by reactions initiated by non-capture processes (processes in which the proton loses energy to the nucleus but is not retained by the nucleus), the reactions for the formation of Fe^{52} and Mn^{52} should perhaps be written $\text{Cu}^{63}(\text{p}, \text{pp}6\text{n}\alpha)\text{Fe}^{52}$ and $\text{Cu}^{63}(\text{p}, \text{p}3\text{n}2\alpha)\text{Mn}^{52}$. On this basis, the yields indicate that the probability of emission of two alpha particles plus three neutrons is much higher than that for the emission of one alpha particle, six neutrons and a proton. Although this effect shows up only in special cases like this where such direct comparisons can be made, it is probably a general phenomenon.

It is also possible on the basis of the spallation yields to say something about the probability of exchange reactions, for example, reactions in which the incident proton undergoes an electron exchange with a neutron in the copper nucleus and emerges as a neutron. The high yields of copper relative to the corresponding zinc isotopes indicate that these exchange reactions do not predominate.

The decreasing yields of individual nuclei far removed from the target nuclei is of course due in part to statistical considerations, and the greater number of possibilities. A larger number of different combinations of emitted particles is possible from a highly excited nucleus than from a nucleus excited to a smaller extent. Thus, even if the probabilities of exciting a nucleus to say 200 and 100 Mev were the same, the yield of an individual product resulting from the higher excitation would be lower than one resulting from the lower excitation.

C. Chemical Separation Procedures

The irradiated copper foil was dissolved in hot concentrated nitric acid or hydrochloric acid and hydrogen peroxide, and milligram amounts of the elements calcium through zinc were then added to act as carrier for the elemental fractions. Known amounts of the elemental fractions to be separated were added so that a quantitative estimation of the amounts of the original carrier lost during the chemical separation procedures could be made in the case of the proton bombardment and the macro amounts of the other elements were added to act as holdback carriers. The chemical separation procedures included distillation, extraction, and precipitation operations.

In general the degree of radiochemical purification required depends on the relative spallation yield and the counting efficiencies of the radio-nuclides in the elemental fraction. If the nuclides are formed in high yield and have a high counting efficiency, the degree of radiochemical purification need not be too high, but if the spallation yields are low or the counting efficiencies are low, the degree of purification must be high.

Usually the particular elements to be investigated were separated successively from the entire dissolved target solution. Since different combinations of elements were removed in each investigation, the chemical separation procedures as a whole varied from bombardment to bombardment. The essential steps for the separation and purification of the elemental fractions were the same and the over-all procedures differed only in the order in which the separations were used. For this reason only the important steps necessary for the chemical separation and purification of

the various elemental fractions are discussed. The final precipitates, if the final step in the chemical separation procedure is a precipitation, were usually dissolved, and aliquots were taken from the solution for counting.

Zinc.-- The dissolved target solution with carriers added was adjusted to 2N in hydrochloric acid and the copper precipitated as the sulfide. The hydrogen sulfide was expelled and the solution neutralized with ammonium hydroxide. The solution was then adjusted to pH 3 with dilute sulfuric acid, and the zinc was then precipitated as the sulfide. The precipitate was dissolved in dilute hydrochloric acid and the hydrogen sulfide expelled by boiling. Three milligrams of iron carrier was added and the solution adjusted to 1N in sodium hydroxide. The ferric hydroxide was removed by centrifugation, and the ferric hydroxide precipitation was repeated. The solution was then adjusted to 1N in hydrochloric acid and the zinc precipitated by adding 4 ml of ammonium mercuric thiocyanate. The zinc mercuric thiocyanate precipitate was washed with a solution containing ammonium mercuric thiocyanate and then transferred to an aluminum plate to be dried and weighed in this form.

Copper.-- The dissolved target solution with carriers added was adjusted to 2N in hydrochloric acid and the copper precipitated as the sulfide. The sulfide precipitate was dissolved in hot concentrated nitric acid, the sulfide expelled or oxidized, and the nitrate destroyed by boiling with concentrated hydrochloric acid. The solution containing the copper was adjusted to 0.5N in hydrochloric acid, sulfur dioxide bubbled through the solution to reduce the copper, and then sodium thiocyanate added to precipitate copper as the cuprous thiocyanate. The cuprous

thiocyanate was washed with water and then dissolved in nitric acid. The nitrate was again destroyed by boiling with concentrated hydrochloric acid and the solution again adjusted to 0.5N in hydrochloric acid. The copper was then precipitated in the final form as the cuprous thiocyanate.

Nickel.— The solution remaining, after the copper had been removed as the sulfide, was boiled to expel the hydrogen sulfide and the iron oxidized with a few drops of nitric acid. The solution was then made alkaline with ammonium hydroxide and the precipitated hydroxides removed. The precipitate was washed with a hot solution of ammonium chloride and the wash combined with the supernate from the precipitation. Three mg of iron carrier was again added and the ferric hydroxide removed by centrifugation. The solution was made slightly acid with acetic acid and 2 ml of a 1 percent alcoholic solution of dimethylglyoxime was added to precipitate the nickel as nickel dimethylglyoxime. The precipitate was washed with water and then dissolved in concentrated hydrochloric acid. One to 2 mg portions of copper, cobalt, and manganese were added to act as holdback carriers, the solution neutralized with ammonium hydroxide, and then made slightly acid with acetic acid. Dimethylglyoxime was again added to make sure that the precipitation of the nickel dimethylglyoxime was complete, and the precipitate was washed again with water. The nickel was weighed as the nickel dimethylglyoxime.

Cobalt.— After the copper had been removed as the sulfide, the solution was boiled to remove the hydrogen sulfide and then neutralized with potassium hydroxide. The solution was adjusted to 3N in acetic acid, and 3 to 4 ml of a saturated solution of potassium nitrite acidified with acetic acid was added to the hot solution containing the cobalt. The

precipitate of potassium cobaltinitrite was allowed to settle in a steam bath, removed by centrifugation, and washed with a 5 percent potassium nitrite solution acidified with acetic acid. The precipitate was dissolved in hydrochloric acid, and holdback carriers for zinc, copper, nickel, and manganese were added. The solution was then neutralized with potassium hydroxide and the cobalt again precipitated as potassium cobaltinitrite. The cobalt was weighed as the potassium cobaltinitrite.

Iron.--- The solution of the copper target, with 5 mg of iron carrier added, was adjusted to 7.75N in hydrochloric acid and the iron extracted with isopropyl ether. The ether layer was washed four times with 6 ml portions of 7.75N hydrochloric acid, and the iron was extracted from the ether layer with water. The solution was made alkaline with ammonium hydroxide and the ferric hydroxide precipitate separated by centrifugation. The precipitate was dissolved in hydrochloric acid and the iron again extracted from 7.75N hydrochloric acid with isopropyl ether. The ether layer was again washed with 7.75N hydrochloric acid and the iron extracted from the ether layer with water. The iron was analyzed by a colorimetric method.

Manganese.--- The solution of the target, to which 5 mg of manganese carrier and holdback carriers for the other elements had been added, was adjusted to 2N in hydrochloric acid and the copper removed as the sulfide. The supernate was made alkaline with ammonium hydroxide and the alkaline sulfides precipitated. The sulfide precipitate was dissolved in concentrated nitric acid and fuming nitric acid added to make the volume up to about 5 ml. Two or three crystals of potassium chlorate were added, and the solution was boiled gently for a few minutes to precipitate manganese

dioxide. The precipitate was removed by centrifugation, washed with water, and then dissolved in one drop nitric acid and one drop of hydrogen peroxide. Holdback carriers for zinc, copper, nickel, cobalt, iron, and chromium were added and the manganese was again precipitated as manganese dioxide from concentrated nitric acid. The manganese was analyzed by weighing as manganese dioxide.

Chromium.-- The steps for the chemical separation of chromium were the same as those given for the separation of the manganese down to the point where the manganese was first removed as manganese dioxide. At this point the chromium was in the form of the chromate and remained in the supernate after the manganese dioxide was removed by centrifugation. The nitric acid solution containing the chromate was adjusted to 0.2N in nitric acid and the solution cooled in an ice bath. Two to three drops of 30 percent hydrogen peroxide were added to form the blue peroxychromic acid, and the peroxychromic acid was extracted with diethyl ether. The ether layer was washed twice with 5 ml portions of water containing a drop of nitric acid. The peroxychromic acid was removed from the ether layer by adding a solution of 0.5N sodium hydroxide which breaks down the peroxychromic acid and forms the chromate. The ether was removed by evaporation, and the excess hydrogen peroxide was destroyed by boiling the solution. The solution was made slightly acid with acetic acid, and the barium was precipitated as the chromate by adding barium chloride dropwise to the hot solution of the sodium chromate. The chromium was weighed as barium chromate.

Vanadium.-- The vanadium was usually separated just subsequent to the extraction of the chromium as the blue peroxychromic acid as described in

the preceding section on chromium. The solution containing the vanadium was almost neutralized with sodium hydroxide and then was poured into 10 ml of a hot solution of 1N sodium hydroxide. The hydroxide precipitate was removed by centrifugation and then washed with 4 ml of hot 1N sodium hydroxide. Two mg each of iron and titanium carrier were then added to the solution and the hydroxides precipitated. The supernatant solution containing the vanadium was boiled to reduce the volume and then made slightly acid with acetic acid. Lead acetate was added to precipitate the vanadium as lead vanadate, and the precipitate was washed with a dilute solution of lead acetate made slightly acid with acetic acid. The lead vanadate was dissolved in nitric acid and the solution adjusted to 2N in nitric acid. The lead was precipitated as the sulfide, and the sulfide was expelled by boiling the solution. Five mg of chromium carrier was added and sulfur dioxide was passed through the solution to reduce the vanadium and chromium. The solution was almost neutralized with sodium hydroxide and then poured into a boiling solution of 1N sodium hydroxide to precipitate the chromium hydroxide. The chromium hydroxide was removed by centrifugation, and the vanadyl ion was oxidized to the vanadate state with hydrogen peroxide. Vanadium was then precipitated as lead vanadate from a solution slightly acid with acetic acid. The vanadium was analyzed colorimetrically.

Titanium.-- After the copper had been removed as the sulfide, the solution with the carriers added was boiled to remove the hydrogen sulfide, and the solution was made alkaline with ammonium hydroxide to precipitate the hydroxides of iron, scandium, and titanium. The precipitate was washed with hot, dilute ammonium hydroxide and then dissolved in hydrochloric acid. The solution was then adjusted to 0.5N in hydrochloric acid and scandium precipitated as the fluoride by adding 0.3 ml of 27N hydrofluoric acid.

After allowing the precipitate to settle for ten minutes on a steam bath, the scandium fluoride was removed by centrifugation, and the supernatant was made alkaline with ammonium hydroxide. The combined titanium and ferric hydroxide precipitate was removed, dissolved in nitric acid and adjusted to 1.6N in nitric acid. Five ml of a solution of potassium iodate was added and the titanium separated as potassium titanium iodate. The precipitate was washed with a dilute solution of the potassium iodate. The iodate was destroyed by bubbling sulfur dioxide into the precipitate mixed with a few ml of dilute hydrochloric acid. Ammonium hydroxide was added to precipitate the titanium hydroxide, the precipitate was removed by centrifugation and then dissolved in hydrochloric acid. Five mg of scandium carrier was added, and the scandium was precipitated as scandium fluoride from a 0.5N solution of hydrochloric acid. The titanium was again precipitated as the hydroxide and the precipitate dissolved in nitric acid. The solution was again adjusted to 1.6N in nitric acid and the potassium titanium iodate again precipitated. The titanium was analyzed colorimetrically.

Scandium.--- The steps in the separation procedure for scandium are the same as those for titanium down to the point where the scandium fluoride is removed. The fluoride precipitate was washed with 0.5N hydrochloric acid containing one drop of 27N hydrofluoric acid. Sulfuric acid was used to dissolve the scandium fluoride and the fluoride ion was removed by fuming the scandium solution with a small amount of concentrated sulfuric acid. The sulfuric acid solution was made alkaline with ammonium hydroxide and the scandium hydroxide precipitate removed by centrifugation. The hydroxide was dissolved in hydrochloric acid and the solution adjusted to 0.5N in hydrochloric acid. Scandium fluoride was then precipitated by adding

0.3 ml of 27N hydrofluoric acid, and the precipitate was allowed to settle on a steam bath. The precipitate was again dissolved in sulfuric acid, the hydrogen fluoride distilled, and scandium hydroxide precipitated from an alkaline ammonium hydroxide solution. The scandium was weighed as scandium oxide.

Calcium.-- After the copper had been removed, the solution containing 5 mg of calcium carrier and holdback carriers for the other elements was made alkaline with ammonium hydroxide, and the alkaline sulfides and hydroxides were precipitated. Holdback carriers for zinc through scandium were again added, and the alkaline sulfides and hydroxides were again precipitated. This last step was again repeated, and the solution was then boiled to remove the hydrogen sulfide. The solution was made slightly acid with oxalic acid, and 5 ml of 4 percent ammonium oxalate was added to precipitate calcium oxalate. The precipitate was allowed to settle on a steam bath for ten minutes, was removed by centrifugation, and washed with water containing oxalic acid and ammonium oxalate. The calcium oxalate was dissolved in concentrated nitric acid, and a few crystals of potassium chlorate were added to oxidize the oxalate ions. Three mg of iron carrier was added, and the solution made alkaline with ammonium hydroxide. The ferric hydroxide was removed by centrifugation and the solution made acid with oxalic acid. Four ml of a solution 4 percent in ammonium oxalate was added to precipitate calcium oxalate as the final purification step, and the precipitate was allowed to settle on a steam bath. The calcium was weighed as calcium oxalate.

Chlorine.-- The copper was dissolved in nitric acid and 5 mg of chlorine in the form of sodium chloride was added. The solution was boiled to distill the hydrogen chloride and the gas was caught in a solution containing

two to three drops of nitric acid and sufficient silver nitrate to precipitate the chlorine as silver chloride. The solution was heated almost to boiling to coagulate the precipitate, and it was removed by centrifugation. The precipitate was washed with dilute nitric acid and then dissolved in dilute ammonium hydroxide. The solution was then acidified with nitric acid and additional silver nitrate added to make sure that the precipitation of the silver chloride was complete. The chlorine was weighed as silver chloride.

Phosphorous.-- The copper target was dissolved in nitric acid and 5 mg of phosphorous as phosphate ion was added to the solution. Holdback carriers were also added for the other elements zinc through calcium. The solution was adjusted to 1N in nitric acid and 10 ml of ammonium molybdate was added to the warmed solution to precipitate ammonium phosphomolybdate. The precipitate was allowed to settle for 15 minutes, removed by centrifugation, and washed with a solution of 1 percent nitric acid. The precipitate was dissolved in ammonium hydroxide containing citrate ions to complex titanium that interferes. Hydrochloric acid was added until the precipitate that formed dissolved with difficulty and 5 mg each of vanadium and titanium carrier were added. Sulfur dioxide was bubbled through the solution to reduce all the vanadium to the vanadyl state. Two ml of cold magnesium chloride was added, and the solution was allowed to stand for five minutes. Concentrated ammonium hydroxide was then added equal to one quarter of the original volume and the precipitate was allowed to settle for 15 minutes. The precipitate was removed by centrifugation and then washed with 3N ammonium hydroxide. The precipitate was dissolved in 1N nitric acid, and the phosphorous was again precipitated as the ammonium phosphomolybdate. The phosphorous was

weighed as the ammonium phosphomolybdate.

Sodium.-- The copper was dissolved in concentrated hydrochloric acid and hydrogen peroxide, and 5 mg of sodium chloride was added as carrier. Carriers for the elements zinc through potassium were added and the solution adjusted to 2N in hydrochloric acid and the copper precipitated as the sulfide. The solution was boiled to dryness to remove the excess acid, the residue dissolved, and then the solution made alkaline with ammonium hydroxide. Hydrogen sulfide was added and the sulfides and hydroxides were precipitated. Additional 3 mg portions of carriers zinc through scandium were added and precipitated. This scavenging process was repeated twice. Excess hydrochloric acid was added to the alkaline sulfide solution, the acidified solution boiled to dryness, and the ammonium chloride driven off. The residue was dissolved in water, the solution checked to make sure it was neutral, and the sodium precipitated as the sodium zinc uranyl acetate. Two 5 ml portions of zinc uranyl acetate were used to wash the precipitate, and the precipitate was then dissolved in absolute alcohol saturated with hydrogen chloride gas. The resulting mixture was cooled in an ice bath and the sodium chloride separated by centrifugation. The sodium chloride precipitate was washed with a 5 ml portion of the alcohol-hydrogen chloride solution. The sodium chloride was dissolved in water and the solution neutralized with potassium hydroxide. Zinc uranyl acetate was added to precipitate the sodium, the precipitate removed by centrifugation, and then washed with additional zinc uranyl acetate. The precipitate was dissolved in absolute alcohol saturated with hydrogen chloride, the mixture cooled in an ice bath, and the sodium chloride removed by centrifugation. The sodium chloride was washed with an

additional portion of the ethyl alcohol-hydrogen chloride solution. The sodium was weighed as sodium chloride.

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LIST OF ILLUSTRATIONS

FIGURE

1. Cross section for formation of Cl^{38} and Na^{24} from copper vs. energy of the bombarding proton.
2. Cross section for formation of Na^{24} from tin vs. energy of the bombarding proton.
3. Aluminum absorption curve for gallium fraction from tin bombarded with 180-Mev protons.
4. Decay of calcium fraction separated from copper irradiated with 340-Mev protons.
5. Decay of calcium fraction separated from copper irradiated with 340-Mev protons counted through sufficient absorber to cut out the beta particles of Ca^{45} and the Sc^{47} daughter.
6. Experimental yields from copper irradiated with 340-Mev protons. Yields are relative to Cu^{61} .
7. Experimental, interpolated, and extrapolated yields from copper irradiated with 340-Mev protons. Yields are relative to Cu^{61} .

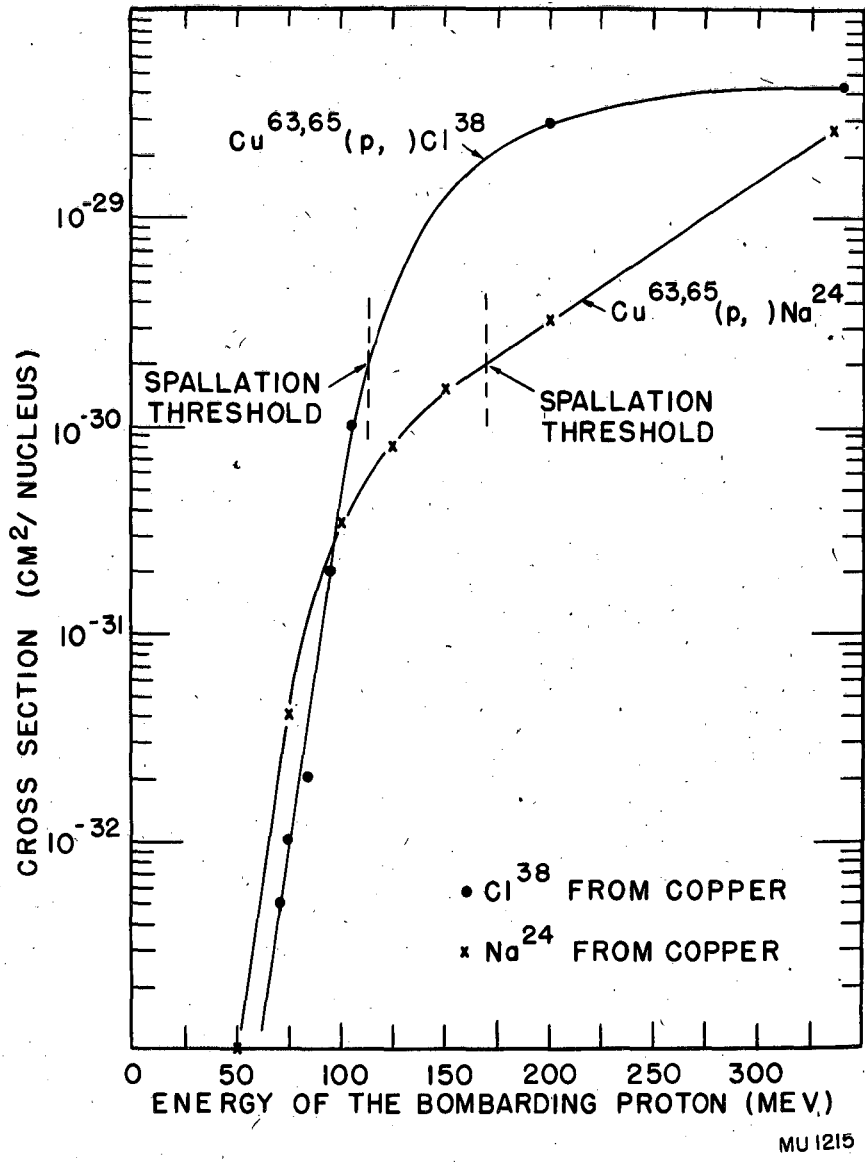


Fig. 1

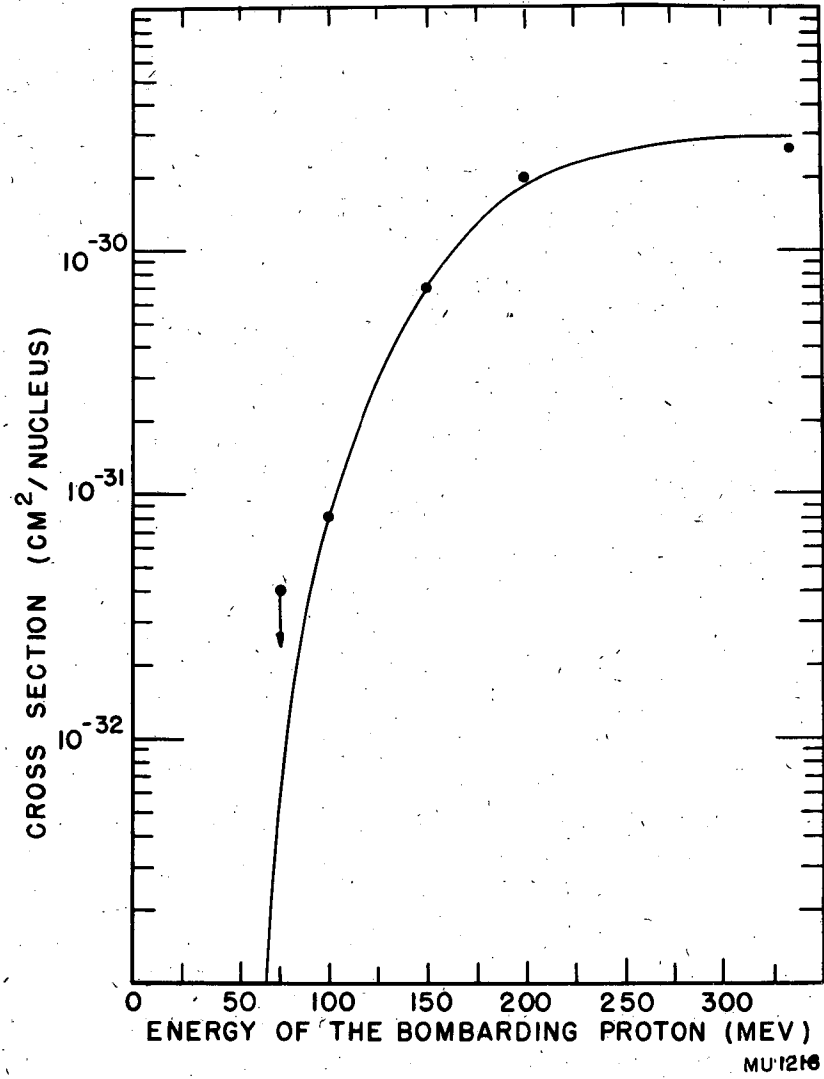


Fig. 2

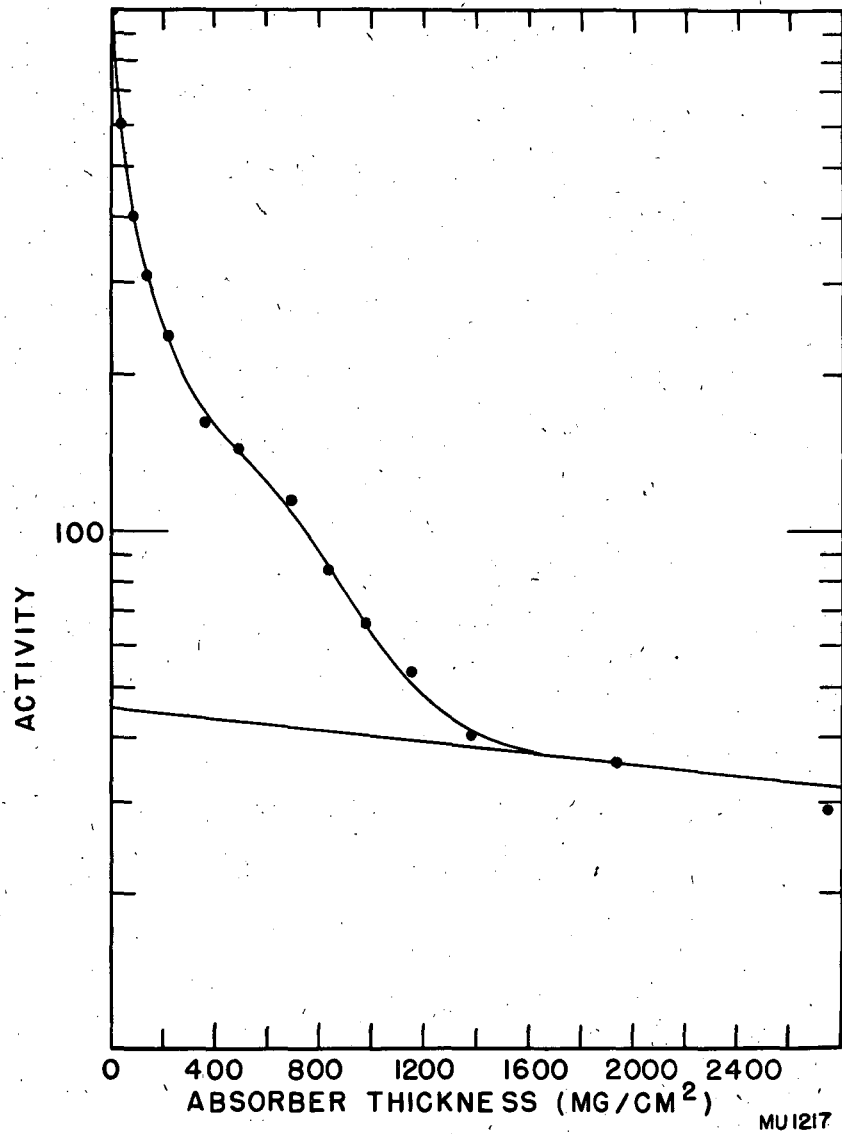


Fig. 3

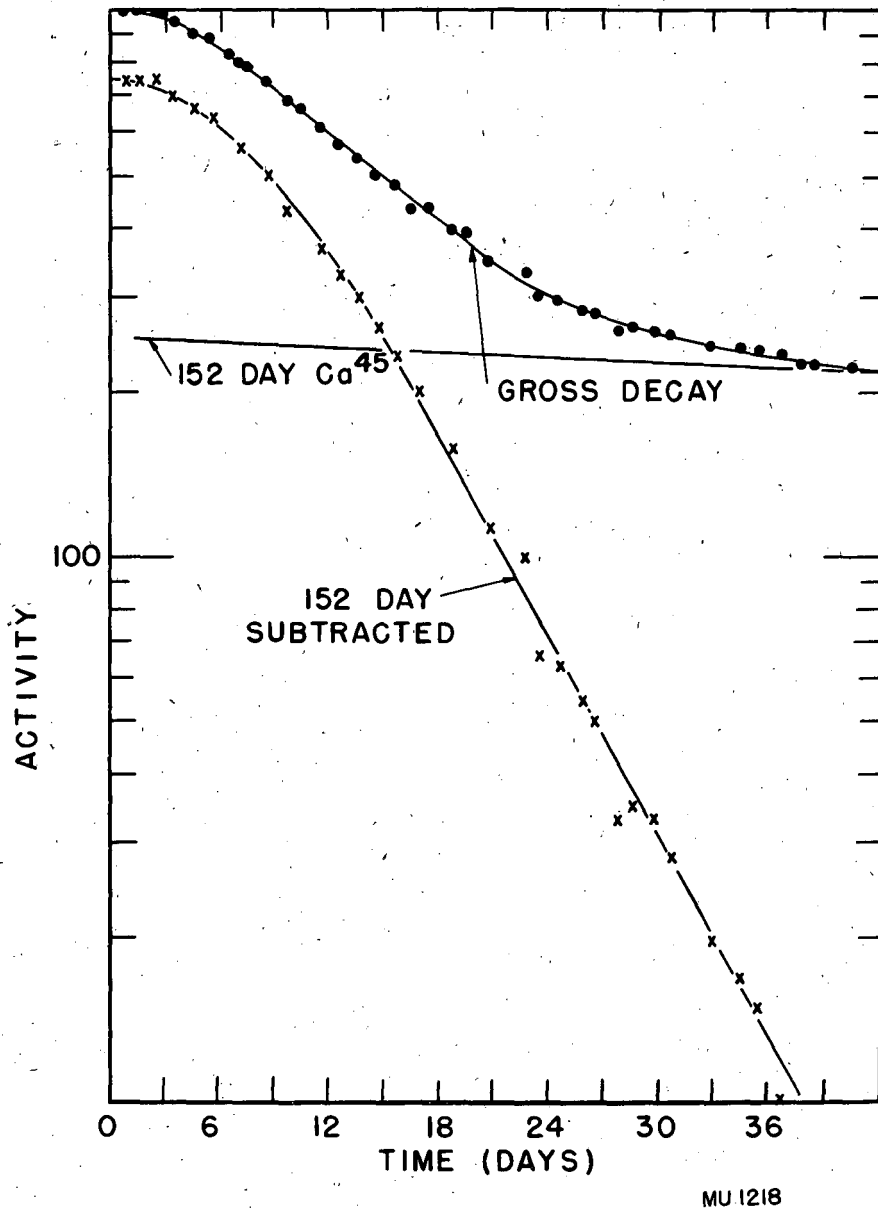


Fig. 4

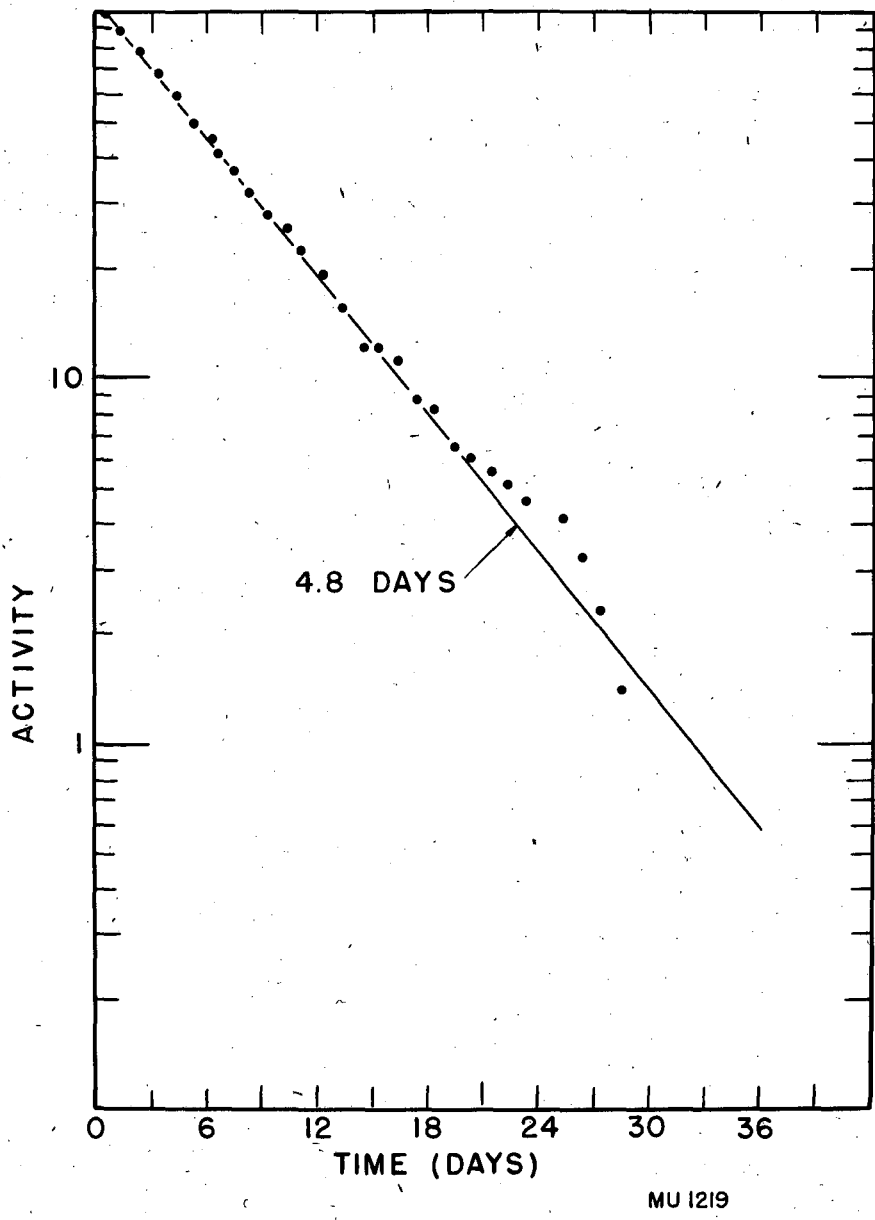
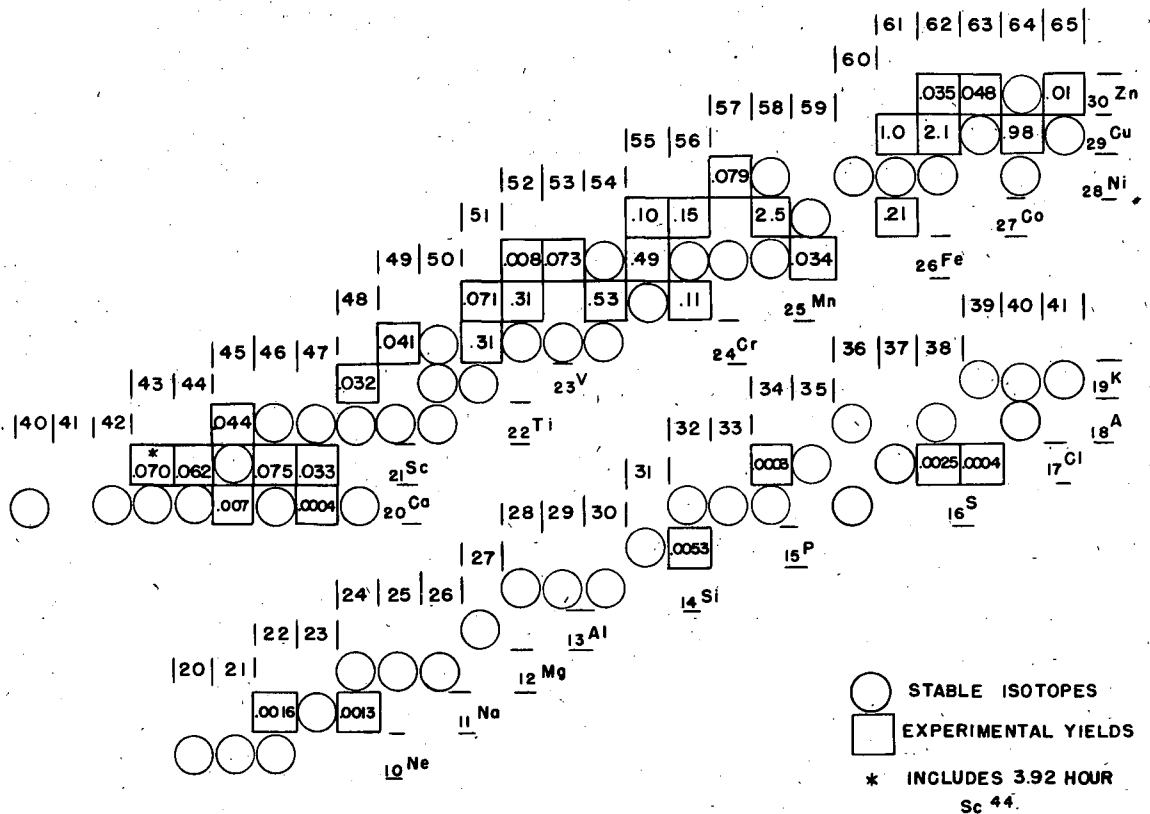
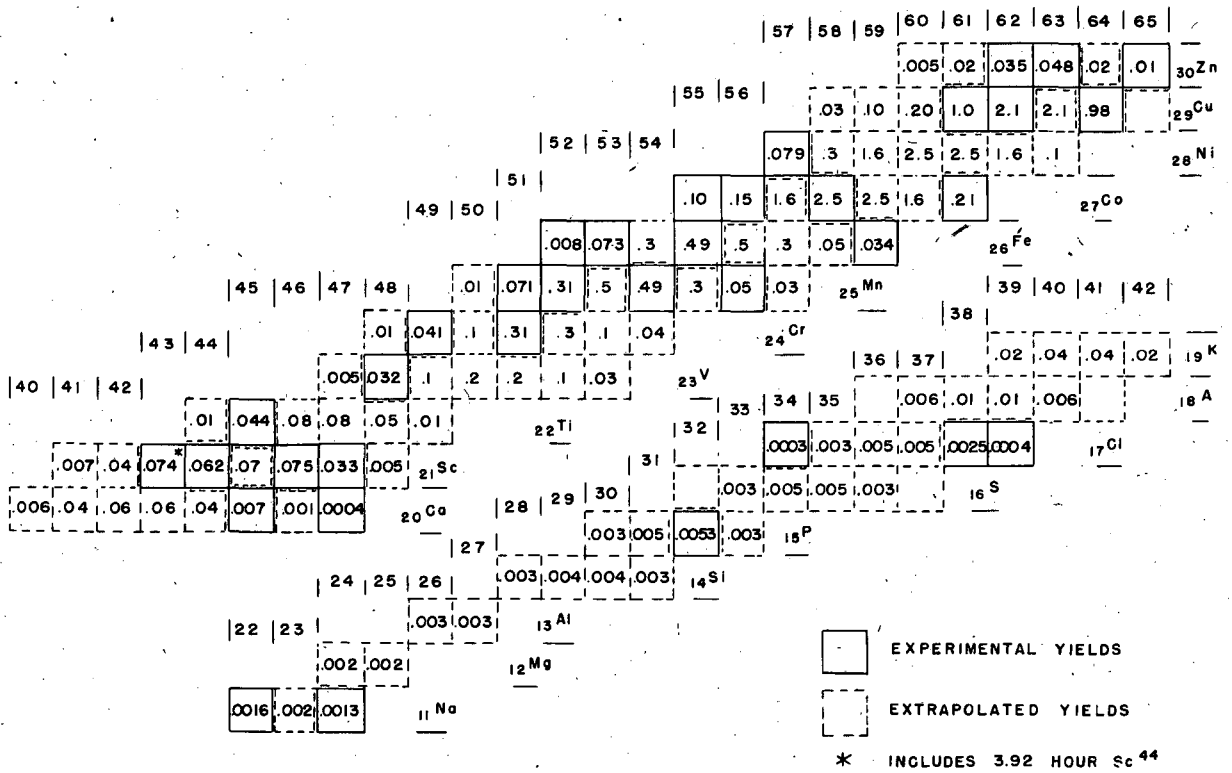


Fig. 5



MU 1220

Fig. 6



MU1223

Fig. 7