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HIGH ENERGY SPALLATION PRODUCTS OF ZINC

William Jacob Worthington, Jr.
(Thesis)

January 8, 1952

Berkeley, California
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HIGH ENERGY SPALLATION PRODUCTS OF ZINC

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ABSTRACT

Elemental zinc was bombarded with 340 Mev protons. A study was made of the various radioactive spallation product fractions resulting from such a bombardment. In the course of the work thirty-four radioactive nuclides from gallium through sodium were identified by separating the various elemental fractions chemically and characterizing the half-lives, type and energy of particulate radiation, and energy of x-rays of the isotopes. The formation cross sections were calculated for the isotopes identified. The general distribution of the spallation products in regard to quantity produced and position in the periodic chart was found to be in general agreement with results previously reported for spallation products of other elements.

One previously unidentified isotope was discovered. This isotope was identified as Ni^{56}.  

*Major, United States Air Force.
HIGH ENERGY SPALLATION PRODUCTS OF ZINC

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I. INTRODUCTION

The term "spallation" has been coined to describe the type of reaction in which the excitation of a nucleus caused by bombardment with small charged particles is degraded by the emission of small nuclear fragments such as neutrons, protons, deuterons, and alpha particles.\(^1\)

The results of a considerable amount of work on spallation reactions have previously been reported.\(^2-8\) The present work consists of the study and determination of radioactive products formed by bombarding elemental zinc with 340 Mev protons. All of the bombardment work was done on the 184-inch cyclotron at the University of California at Berkeley, California.

Some thirty-four nuclides from sodium through gallium were observed as spallation products in the course of the present work. The general distribution of these products in regard to quantity produced and position in the periodic chart is in agreement with results previously reported for similar reactions on other elements such as arsenic,\(^8\) antimony,\(^3\) copper,\(^7\) iron,\(^5\) and nickel.\(^6\)

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II. PROCEDURE

A. Description of Targets and Method of Monitoring Beam

Irradiations with 340 Mev protons were carried out in the circulating beam of the 184-inch frequency modulated cyclotron. The targets were of two types, a thin target, and a thick target. A thin target consisted of a 5 mil spectroscopically pure zinc foil between two 1 mil aluminum guard foils, all three of which were sandwiched between two 1/2 mil aluminum monitor foils (see Figure 1). These thin foils which were about 1 1/4 inches long and 3/8 inch wide were clamped in target holders which could be attached to the movable probe head of the cyclotron. The bombarding energy was adjusted by fixing the radial distance of the leading edge of the target strip from the origin of the beam. Since the degradation of the energy of the high energy particles in passing through the 3 mils of aluminum foil and the 5 mils of zinc foil is small, such a target could be considered as a thin target. The thick target consisted of six 5 mil zinc foils approximately 1 1/2 inches long and 3/4 inch wide clamped together and held in an envelope of 1 mil aluminum foil. This was bombarded in like manner to the thin target except that the target was so arranged that the proton beam struck the edge of the foils and passed through the 3/4 inch width of the foils (see Figure 2).

The exact amount of irradiation of the zinc target could not be determined directly because of the fluctuation in intensity of the circulating beam. The thin targets were trimmed along the leading edge and side after being placed in the target holder so that the edges of all
the foils were exactly aligned. After bombarding and while the foils were still clamped in the holder, the foils were simultaneously cut off on a chopping block so that the bombarded area of both the zinc target foil and the two aluminum monitor foils were identical. The aluminum monitor foils were later cut and attached to scotch tape in such a manner that their area and configuration approximated that of the sample as counted for each of the various spallation product fractions taken from the zinc (see Figure 3B). The monitor foils were weighed before counting, and the monitor foils and spallation product fraction were counted under identical geometry conditions in the counter. Decay of Na$^{24}$ in the aluminum monitor foils was followed on the counter. The weights and disintegration rate of the two monitor foils were averaged and the arithmetical mean of the two was used in all calculations. The cross section for the formation of Na$^{24}$ from aluminum under bombardment with 340 Mev protons was taken as 10 mb. The cross sections for the formation of the various spallation products from zinc were calculated relative to the cross section for the formation of Na$^{24}$ from aluminum. The zinc and aluminum monitor foils were of identical area and were bombarded under the same conditions. The monitor foils and the various spallation fractions were counted under virtually identical conditions, and the absolute cross section for the Na$^{24}$ was known. It was therefore possible to calculate easily the absolute cross sections for formation of the various spallation products from the weight of the aluminum monitor foils, the weight of the zinc foil, the respective disintegration constants, the duration of bombardment, and the observed counting rate.
The absolute cross section for the formation of Cu\textsuperscript{67} from zinc was determined four times using the above procedure. The results of three of these determinations checked very well. In a few of the thin targets where difficulties were encountered in accurately cutting the foils, the cross sections of the spallation products determined were calculated relative to the cross section for formation of Cu\textsuperscript{67} (4.7 mb). The absolute cross section for formation of Cr\textsuperscript{51} was likewise determined in thin targets. In the thick targets, where aluminum monitor foils could not be used, the cross sections of the spallation products determined were calculated relative to the cross section for formation of Cr\textsuperscript{51} (15 mb).

After the target was bombarded, it was dissolved in an acid solution containing measured amounts of inactive carriers. The various elemental fractions were separated by chemical separation procedures described below. The amount of carrier recovered after the chemical separation of an elemental fraction was determined gravimetrically; the percent of the carrier recovered was assumed to be the same as the percent of the radioactive atoms of the element recovered.

B. Counting Procedures

Counting of the various activities was done on an end window, chlorine quenched, argon filled, Geiger counter tube (Amperex type 100-C) with a mica window of approximately 3 mg/cm\textsuperscript{2} thickness. Combined air and window thickness was approximately equivalent to 5.8 mg/cm\textsuperscript{2} of aluminum from shelf 2 of the counter and 11.5 mg/cm\textsuperscript{2} of aluminum from shelf 5 of the counter. A counting scale of 256 was generally used. The various
isotopes were characterized by half-life determinations, absorption measurements, and type of particulate radiation emitted. The type of particulate radiation emitted was determined by use of a crude beta ray spectrometer.

The counting data were corrected for air and window absorption, coincidence, background, backscattering, self-scattering and self-absorption, and, where x-rays were counted, for fluorescent yields of the x-rays. Counting efficiencies of x-rays in argon were taken as those determined by Studier and James, while backscattering factors were those determined by Yaffe and Justus. Self-scattering and self-absorption factors were taken from the work of Nervik and fluorescent yields, from the data of Compton and Allison. It was assumed that all the particulate radiation which entered the Geiger tube counted with the same efficiency regardless of the energy or sign of the radiation.

The mountings used were varied. In some cases the samples were mounted on 4 cm² 1 mil aluminum plates which were then mounted on either scotch tape or on a plastic plate of approximately 92 mg/cm² thickness. In other cases they were mounted on 1 cm² 1 mil aluminum plates which were then mounted on saturation backscattering aluminum plates (see Figure 3). Coincidence factors were determined empirically by following the decay of a pure Y²⁹ sample on all shelves from an activity of approximately 150,000 cpm down to approximately 1000 cpm. By back extrapolating the decay curve mathematically, the coincidence at different counting rates was determined as the difference between the calculated rate of decay and the rate as measured on the counter. In addition to all other
corrections, an empirical correction factor was used to take into account the fluctuation in counting efficiency of the Geiger tube. A sample of $^{238}U$ was counted once or twice a day and all counts of various spallation products were corrected to an arbitrarily assumed standard counting rate for this $^{238}U$ sample. The maximum deviation in the standard was approximately 3 percent from the arbitrarily chosen standard counting rate. Generally the fluctuation was less than 1 percent.

Air and window absorption was determined either by straight line extrapolation of aluminum absorption curves or mathematically from the known air and window thickness and the aluminum half thickness of the particulate radiation.

C. Chemical Separation Procedures

The irradiated zinc foil was first weighed and then dissolved in a solution containing concentrated nitric acid or hydrochloric acid along with a known amount (usually 3 to 10 milligrams) of inactive carrier for each of the elemental fractions to be separated. In addition inactive carriers for the other elements gallium through potassium (except zinc) were generally added to act as holdback carriers for the elemental fractions which were not to be separated.

The individual elements to be investigated in each bombardment were successively separated from the entire target solution (except in the case of zinc where an aliquot of the target solution was taken). These elemental fractions were then purified, precipitated, dried or ignited to known composition and weighed. The percent recovery of the known
amount of inactive carrier originally added was assumed to be the same as the percent recovery of the radioactive atoms of the particular element under investigation. The degree of purification required depended upon the relative cross section for formation and upon the half-life of the isotope being investigated. An isotope with a low cross section for formation required more intensive purification than did one with a high cross section. Likewise an isotope with long half-life required more intensive purification than did one with a short half-life.

The principal steps in preparation and purification of the individual elemental fractions were generally the same in all bombardments. Since different combinations of elements were recovered from different targets, however, the order in which these steps were taken varied from bombardment to bombardment. Minor modifications in the procedure were used from time to time where necessary.

The important steps necessary for separation and purification of the various elemental fractions are discussed below.

**Gallium.** The target was dissolved in hydrochloric acid and the solution adjusted to 7.5 N in hydrochloric acid. The solution was intimately mixed with successive portions of isopropyl ether saturated with hydrochloric acid. The ether layer containing the gallium and iron fractions was drawn off after each mixing and finally all ether layers were combined and washed three times with 7.5 N HCl. The gallium and iron were then re-extracted into water in successive steps. The water layers were combined and the iron reduced with stannous chloride. The solution
was again adjusted to \(7.5 \text{ N}\) with respect to hydrochloric acid and the gallium was extracted into isopropyl ether saturated with hydrochloric acid. The iron in the ferrous state remained behind in the aqueous layer during this extraction. Ether layers were combined and washed with \(7.5 \text{ N} \ \text{HCl}\). The gallium was then re-extracted into water. Approximately 2 mg of inactive iron was added to the solution as ferric ion and the solution adjusted to at least 1 N with sodium hydroxide. The ferric hydroxide was centrifuged off and discarded. The ferric hydroxide scavenge was repeated, the solution was then adjusted to 0.5 to 1 N in hydrochloric acid, one or two milligrams of copper carrier was added, and the copper was precipitated as the sulfide with hydrogen sulfide gas. The copper sulfide was removed and the solution was heated to remove the \(\text{H}_2\text{S}\). The solution was then adjusted to \(\text{pH 5.5}\) with ammonium hydroxide using brom cresol purple as an indicator. An equal volume of \(\text{pH 5.5}\) buffer solution composed of ammonium acetate and acetic acid was added and the solution allowed to digest for five minutes in a hot water bath. The gallium hydroxide was centrifuged off, washed twice with the \(\text{pH 5.5}\) buffer solution, dried, transferred to a porcelain crucible and ignited in a muffle furnace at 1300° F for 30 minutes. The gallium oxide was dried in a desiccator, transferred to an aluminum counting dish and weighed as \(\text{Ga}_2\text{O}_3\). Due to the fact that the gallium oxide is highly deliquescent, it was necessary to make all weighings in a glass stoppered weighing bottle.

**Zinc.** After all other elements being investigated had been removed from the solution, an aliquot of approximately 2 to 5 percent of the
remaining solution was taken and adjusted to 0.4 N with respect to hydrochloric acid. Inactive copper carrier was added and the copper precipitated as the sulfide with hydrogen sulfide gas. The copper sulfide precipitate was discarded and the solution boiled to remove all hydrogen sulfide. The zinc was precipitated with ammonium mercuric thiocyanate solution and washed twice with water containing 10 ml of the precipitating reagent per liter. The zinc mercuric thiocyanate was then dissolved in concentrated nitric acid, the nitric acid destroyed with formic acid and the solution adjusted to 2 N with respect to hydrochloric acid. The mercury was precipitated from the solution with hydrogen sulfide gas and removed by centrifugation. The solution was boiled to remove all hydrogen sulfide gas and scavenged with ferric hydroxide by adding ferric ion, making it strongly ammoniacal and removing the ferric hydroxide by centrifugation. The solution was carefully neutralized with hydrochloric acid using brom thymol blue as an indicator, and then heated nearly to boiling in a water bath. An excess of \((\text{NH}_4)_2\text{HPO}_4\) reagent was added to the solution which was then digested until the resulting precipitate became crystalline. The precipitate was removed by centrifugation, washed once with the precipitating solution diluted one to ten, twice with water, twice with alcohol and finally with ether. The precipitate was dried at 120° C for 30 minutes and weighed as \(\text{Zn(NH}_4\text{P}_4\).}

**Copper.** The solution was made ammoniacal and the copper was reduced with \(\text{Na}_2\text{SO}_3\). Potassium thiocyanate was added to the solution and cuprous thiocyanate was precipitated by making the solution acid with acetic acid.
The precipitate was removed by centrifugation and washed twice with water. The cuprous thiocyanate was dissolved in a minimum amount of concentrated nitric acid and diluted to approximately 7 cc. A drop of hydrochloric acid was added and the solution scavenged twice with silver nitrate. Ferric ion was added and the solution scavenged by bringing down ferric hydroxide with ammonium hydroxide. The ferric hydroxide scavenging was repeated. The copper was reduced with Na₂SO₃. Potassium thiocyanate was added to the solution, and cuprous thiocyanate was again precipitated when the solution was acidified with acetic acid. The precipitate was centrifuged off, washed twice with water, twice with alcohol, dried at 110° C for 30 minutes and weighed as CuCNS.

**Nickel.** The solution was twice scavenged by precipitating ferric hydroxide from an ammoniacal solution. A saturated alcoholic solution of dimethyl glyoxime was added in excess and the nickel removed as nickel dimethyl glyoxime by centrifugation. The precipitate was washed twice with water and dissolved in a small amount of concentrated nitric acid. The solution was diluted and scavenged twice by precipitating silver chloride. The solution was again made ammoniacal, and the nickel was precipitated as the dimethyl glyoxime. The precipitate was washed twice with water, twice with alcohol, dried at 115° C for 30 minutes and weighed as C₈H₁₄N₄O₄Ni.

In those cases where it was desired to make sure the nickel was free of cobalt, the nickel dimethyl glyoxime precipitate was dissolved in a small amount of concentrated hydrochloric acid, cobalt hold-back carrier
was added, and the solution was run through a Dowex A-2 resin column. The cobalt remained upon the column, and the nickel did not. An excellent separation could be made in this way. The nickel from one of the thin targets (bombardment number 2) and from both of the thick targets (bombardments number 11 and 12) were purified in this manner. The nickel in the solution run through the column was precipitated as the dimethyl glyoxime from an ammoniacal solution. In bombardments number 11 and 12, a second column separation was made, a known amount of cobalt carrier being added prior to the last column run and later recovered and checked for activity.

Cobalt. After other hydroxides were removed from an ammoniacal solution, the solution was acidified and the cobalt then precipitated as the hydroxide with strong sodium hydroxide solution. The cobalt hydroxide was removed by centrifugation and washed twice with water. The cobalt was then dissolved in a weak hydrochloric acid solution and complexed with a concentrated solution of KCNS. The cobalt was then extracted into a 1:1 mixture of amyl alcohol and ether, the extraction being repeated until no further blue cobalt complex could be detected. The amyl alcohol-ether extractions were combined and washed with a concentrated water solution of KCNS acidified with hydrochloric acid after which the cobalt was re-extracted into ammonia. The cobalt was precipitated from the ammoniacal solution as the sulfide with hydrogen sulfide gas. The cobalt sulfide was removed by centrifugation, washed with water, and dissolved in a minimum amount of concentrated nitric acid. The solution was diluted
to approximately 8 cc and the sulfur which had formed was removed by centrifugation. The acid solution was neutralized with solid potassium carbonate and made acidic with acetic acid. The cobalt was finally precipitated from hot solution as the potassium cobaltinitrite by the addition of a freshly prepared very concentrated solution of potassium nitride acidified with acetic acid. The precipitate was removed by centrifugation, washed twice with water and twice with alcohol, dried at 120° C for 30 minutes and weighed as K₃Co(NO₂)₆·H₂O.

Iron. The iron was extracted into isopropyl ether along with gallium from a 7.5 N HCl solution. The extraction was washed with 7.5 N HCl and the iron and gallium re-extracted into water. The iron was reduced with stannous chloride, the solution made 7.5 N with respect to hydrochloric acid and washed several times with isopropyl ether to remove gallium. The iron was then oxidized by adding a few drops of nitric acid and heating, and again extracted into isopropyl ether. The ether extractions were washed three times with 7.5 N HCl. The iron was re-extracted into acetic acid-ammonium acetate solution and then precipitated as the hydroxide by making the solution 1 N with respect to sodium hydroxide. The hydroxide was washed twice with hot water and redissolved in 1 N HCl. The solution was scavenged by adding a drop of copper carrier and precipitating the copper as a sulfide with hydrogen sulfide gas. The copper sulfide scavenging was repeated. The solution was boiled to remove the hydrogen sulfide gas and made strongly ammoniacal. The ferric hydroxide was removed by centrifugation, washed twice with hot water, dried,
transferred to a porcelain crucible, and ignited at 900° F for approximately 20 minutes. The iron was then weighed as Fe₂O₃.

**Manganese.** The solution was made approximately 1 N with respect to nitric acid and carriers gallium, copper, nickel, iron, cobalt, chromium, vanadium, titanium, calcium and potassium, and yttrium were added as holdback carriers. Potassium bromate was added to the solution which was then digested in a hot bath until the brown manganese dioxide precipitated. The precipitate was removed and washed with hot water then redissolved in nitric acid and a few drops of hydrogen peroxide. The solution was adjusted to 1 N with respect to nitric acid and heated to remove the hydrogen peroxide. The solution was twice scavenged with silver chloride and one drop each of all carriers gallium through potassium (except for zinc, manganese, and scandium) were added to act as holdback carrier. The manganese was again precipitated as the dioxide with potassium bromate. The precipitate was washed twice with hot water and then dissolved in 0.5 N HCl to which a crystal of oxalic acid had been added. The solution was then scavenged twice by adding copper carrier and precipitating the copper as the sulfide with hydrogen sulfide gas. The solution was made 1 N with respect to nitric acid and the manganese precipitated as the dioxide with KBrO₃. The manganese dioxide was washed twice with hot water and twice with acetone, dried in a hot water bath, powdered, transferred to a porcelain crucible, ignited at 900° F and weighed as Mn₃O₄.
Chromium. The chromium was oxidized to the chromate state (with hydrogen peroxide if in basic solution or with potassium bromate if in acid solution). The solution was neutralized and then made barely acidic with nitric acid. The solution was cooled in an ice bath and then mixed intimately with an equal volume of ice cold ether in a separatory funnel. While the solution was being mixed, a drop of hydrogen peroxide was added. The intensely blue peroxychromic acid was extracted into the ether. The extraction process was repeated until no further peroxychromic acid could be extracted. The combined ether layers were washed three times with ice cold water acidified with a drop of nitric acid. The chromium was then extracted into dilute potassium hydroxide solution. The solution was made barely acid with nitric acid and cooled. The extraction process was repeated in its entirety. After extracting the second time into dilute potassium hydroxide solution, the chromium solution was made acidic with acetic acid, and holdback carriers of the elements gallium through potassium (with the exception of zinc and scandium) plus yttrium holdback carrier were added. Barium acetate was added to the solution causing barium chromate to precipitate. The \( \text{BaCrO}_4 \) was centrifuged off, washed twice with water and dissolved in a minimum amount of hydrochloric acid. The solution was diluted to approximately 10 cc and ammonium acetate solution was added until the barium chromate reprecipitated. The barium chromate was centrifuged off, washed twice with water, and twice with alcohol, dried at 115°C for 15 minutes and weighed as \( \text{BaCrO}_4 \).

Vanadium. The solution containing carriers of all elements from gallium through potassium was made alkaline with solid sodium hydroxide.
The hydroxides were removed by centrifugation and one drop of all carriers gallium through potassium except for vanadium were again added to the solution and the hydroxide scavenge repeated. The solution was then made acidic with acetic acid, a few drops of hydrogen peroxide were added and the solution was heated to oxidize the vanadium to the vanadate state. Under these conditions any chromium in the solution is reduced so that it does not interfere with the later vanadium precipitation. The solution was then boiled to remove the hydrogen peroxide and to reduce the volume to approximately 8 cc. Approximately 5 cc of 10 percent lead acetate solution was then added and the solution was digested on the hot bath. Lead vanadate, which precipitated, was removed by centrifugation and washed twice with dilute lead acetate solution acidified with a few drops of acetic acid. The lead vanadate was then dissolved in a minimum amount of concentrated nitric acid and the solution was diluted to approximately 8 cc. The lead vanadate was reprecipitated from the solution by the addition of a concentrated solution of ammonium acetate. The precipitate was removed by centrifugation, washed twice with a few cc of water and once with absolute alcohol, dried at 115° C for 20 minutes and weighed as Pb(VO3)2.

Titanium. Titanium was precipitated as the hydroxide and washed with water. The hydroxide was dissolved in a minimum amount of nitric acid. Scandium carrier was added to the solution and then a few drops of HF were added to precipitate scandium fluoride. The scandium fluoride was removed and discarded and the scandium scavenge repeated. The solution
was then made ammoniacal, and the titanium hydroxide was removed by centrifugation and washed twice with dilute ammonium hydroxide. The titanium hydroxide was dissolved in 7.5 N HCl, and iron carrier was added to the solution. The solution was then washed several times with isopropyl ether saturated with hydrochloric acid. The titanium was again reprecipitated as the hydroxide with ammonia and washed with dilute ammonia. The titanium hydroxide was dissolved in 2 N HNO₃, and potassium iodate was added to precipitate titanium iodate which was then centrifuged off and washed twice with dilute potassium iodate solution. The titanium iodate was dissolved in 1 N HCl solution containing sodium sulfite. The solution was made basic with ammonia, and the titanium hydroxide was removed by centrifugation. The precipitate was washed twice with water and twice with acetone, dried in a hot bath, transferred to a porcelain crucible, ignited at 1300° F for 20 minutes and weighed as TiO₂.

**Calcium.** The calcium was precipitated as the carbonate by the addition of a strong solution of ammonium carbonate to an ammoniacal solution. The calcium carbonate precipitate was washed twice with water and then dissolved in nitric acid. The solution was scavenged by adding a few drops of yttrium and titanium carrier, making the solution basic with ammonia, and removing the hydroxide which precipitated. Manganese carrier was added, and the solution was scavenged by precipitating manganese dioxide with hydrogen peroxide. Cobalt carrier was added and precipitated as the sulfide with gaseous hydrogen sulfide. The cobalt sulfide scavenge was removed, the solution was heated to remove the hydrogen sulfide
and made acid with acetic acid. A concentrated solution of ammonium oxalate was added to the hot solution which was then digested in a hot bath for a few minutes. The calcium, which precipitated as the oxalate, was washed twice with water and then dissolved in 6 M HNO₃. The oxalate was destroyed by the addition of a little KClO₃ and heating. The solution was then made ammoniacal and scavenged by precipitating ferric hydroxide which was then removed by centrifugation. The solution was acidified with oxalic acid and ammonium oxalate and allowed to digest in a hot bath. The calcium oxalate was removed by centrifugation, washed twice with water and twice with alcohol, dried at 115° C for 30 minutes and weighed as CaC₂O₄·H₂O.

**Potassium.** The solution was made weakly acidic with acetic acid and most of the zinc precipitated and removed as sulfide with gaseous hydrogen sulfide. A drop of copper carrier was added and the sulfide scavenger repeated. The solution was made alkaline with ammonia, a drop each of copper and cobalt carrier were added and the solution was again given a sulfide scavenger. The hydrogen sulfide was removed by boiling and the solution again made acidic with acetic acid. Approximately 4 mg of phosphate carrier was added to the solution and precipitated with a ferric chloride solution. The phosphate scavenger having been removed by centrifugation, the solution was scavenged for hydroxides by adding titanium carrier and precipitating the iron and titanium hydroxides with ammonia. The hydroxide scavenger was repeated. Chromium carrier was added to the solution and precipitated and removed as the sulfide. The
solution was boiled to remove hydrogen sulfide. Calcium carrier was added, precipitated with ammonium carbonate, and removed by centrifugation. The solution was made acid with hydrochloric acid and evaporated to dryness. Ammonium chloride was fumed off over a Meker burner and the organic residue burned off at the same time. The residue was treated with a few cc of water and that portion which did not dissolve was removed by centrifugation. One cc of 72 percent perchloric acid was added, and the solution was heated until fumes of perchloric acid were given off. The solution was cooled in an ice bath. Absolute alcohol plus a few drops of perchloric acid were added and the potassium perchlorate which precipitated was centrifuged off. The precipitate was washed with absolute alcohol and redissolved in approximately 1 cc of water. The potassium perchlorate was reprecipitated by adding absolute alcohol and a few drops of perchloric acid to the solution. The potassium perchlorate was again centrifuged off, washed with alcohol, redissolved in water and once again recrystallized. The final precipitation was washed twice with absolute alcohol, dried at 130° C for one hour and weighed as KClO₄.

Sulfur. After removing hydroxides and calcium from the solution, the sulfur was precipitated as barium sulfate by adding barium chloride to the acid solution. The barium sulfate was removed by centrifugation, washed twice with water and then digested in a hot bath with a few cc of concentrated potassium carbonate solution. The supernatant was removed and the precipitate again heated with concentrated potassium carbonate solution. The supernatant was again removed and added to that previously
recovered. The combined supernatants were carefully neutralized with hydrochloric acid and then made 1 N with hydrochloric acid and heated to remove carbon dioxide. The sulfur was again precipitated as barium sulfate by adding barium chloride to the solution. The barium sulfate was removed, washed twice with water and the metathesis with potassium carbonate repeated in its entirety. The sulfur was finally precipitated as barium sulfate, washed twice with water and twice with alcohol, dried for 15 minutes at 115° C and weighed as BaSO₄.

**Phosphorus.** The solution was made 2 N with respect to nitric acid and the phosphorus precipitated as ammonium phosphomolybdate by adding approximately 5 cc of ammonium molybdate solution¹⁷ and digesting for 10 minutes. The precipitate was removed, washed with dilute nitric acid and dissolved in ammonia and ammonium citrate solution. The solution was rendered slightly acid with hydrochloric acid, and a few drops of vanadium and titanium carrier were added. Sulfur dioxide was bubbled through the solution to reduce the vanadium. Approximately 3 cc of magnesium chloride solution¹⁸ was added to the solution, which was then allowed to stand for 10 minutes. The solution was next neutralized to phenolphthalein end point and then diluted with an additional 25 percent of its volume of concentrated ammonium hydroxide. The solution was allowed to sit for an hour or more, and then the precipitate was removed by centrifugation and washed with 3 N NH₄OH. The precipitate was dissolved in 1 N HNO₃ and the ammonium phosphomolybdate again precipitated and washed with dilute nitric acid. The precipitate was dissolved in
ammonia and hydrochloric acid was added until the precipitate which formed dissolves with difficulty. Two cc of the magnesium chloride solution was added, and the solution was heated in a hot bath and then neutralized to phenolphthalein end point with ammonia. Concentrated ammonium hydroxide in volume equal to 25 percent of the volume of the solution was added and the solution digested at 65° C in a hot bath for 30 minutes. The precipitate was removed by centrifugation, washed twice with water, and twice with alcohol, dried at 105° C for one hour and weighed as Mg(NH₄)PO₄·H₂O.

Sodium. The solution containing holdback carriers of the elements gallium through scandium (except zinc) was scavenged of hydroxides. Gaseous hydrogen sulfide was bubbled through the ammoniacal solution and the sulfides discarded. Calcium carrier was added and precipitated as the carbonate with ammonium carbonate. After removing and discarding the calcium carbonate scavenge, the solution was made acid with acetic acid, copper carrier was added, precipitated as the sulfide with gaseous hydrogen sulfide, and removed by centrifugation. The solution was made basic with ammonia, more copper carrier was added, precipitated as the sulfide, and removed. The solution was made slightly acid with hydrochloric acid and more copper carrier was added, precipitated as the sulfide, and removed. Excess hydrochloric acid was added to the clear solution which was then evaporated to dryness. Ammonium chloride was fumed off over a Meker burner. The residue was treated with approximately 4 cc of water. Insolubles were removed by centrifugation, and to the clear solution an excess of zinc uranyl acetate solution was added causing the sodium to
precipitate as sodium zinc uranyl acetate. This precipitate was removed by centrifugation, washed with the zinc uranyl acetate solution, and dissolved in absolute alcohol saturated with hydrogen chloride gas. The alcoholic solution was cooled in an ice bath. The sodium chloride which precipitated was removed by centrifugation. The sodium chloride was dissolved in approximately 4 cc of water and the resultant solution was neutralized with potassium hydroxide. The sodium was again precipitated as the sodium zinc uranyl acetate and the crystallization of sodium chloride from alcoholic solution was repeated in its entirety. The final precipitate was washed twice with absolute alcohol saturated with hydrogen chloride gas, dried at 115° C for 30 minutes and weighed as NaCl.

III. RESULTS

A. Radioactive Nuclides Identified

The characteristic activities of the isotopes discussed in this section were observed in the elemental fractions separated from the zinc foil targets. References for most of the isotopes may be found in National Bureau of Standards Circular Number 499, "Nuclear Data" of September 1, 1950, as amended by Supplement Number 1, April 25, 1951. Only in cases where information on the isotope was published in the literature subsequent to the data listed in Circular 499 will references be cited.

Gallium Fraction. The activities of Ga\(^{66}\) and Ga\(^{67}\) were observed in the gallium fractions. The 9.4 hour Ga\(^{66}\) was identified by its half-life
and aluminum absorption curve and was assumed to decay 34 percent by electron capture and 66 percent by positron emission. The energy and relative abundance of the positrons emitted is assumed to be as follows:

<table>
<thead>
<tr>
<th>% of Total Positron Decay</th>
<th>Energy (Mev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>87</td>
<td>4.14</td>
</tr>
<tr>
<td>4.3</td>
<td>1.4</td>
</tr>
<tr>
<td>6.9</td>
<td>0.878</td>
</tr>
<tr>
<td>1.7</td>
<td>0.403</td>
</tr>
</tbody>
</table>

The Ga$^{67}$ which decays 100 percent by electron capture was identified by its 78 hour half-life and by the characteristic zinc K x-ray as seen on an aluminum absorption curve where the aluminum absorbers were sandwiched between two beryllium absorbers of 13.8 mg/cm$^2$ thickness each. From a beryllium absorption curve it was determined that 50 percent of the counts registered were due to electromagnetic radiation. From the aluminum and beryllium absorption curve it was determined that of this 50 percent, 90 percent were due to zinc K x-rays. Since the 93 kev gamma ray which accompanies each Ga$^{67}$ disintegration has an internal conversion coefficient of two, there are 5 x-rays from every 3 disintegrations. Only 42 percent of these K x-rays are assumed to escape from the atom. Zinc K x-rays are assumed to have a counting efficiency of 2.7 percent. The yield of Ga$^{67}$ was calculated on the basis of counts of x-rays using the figures as given above.
Zinc Fraction. The activities of Zn$^{62}$ and Zn$^{65}$ were observed in the zinc fractions. The Zn$^{62}$ was identified by means of its half-life and by the sign of its particulate radiation as determined on the crude beta ray spectrometer. It was assumed to decay 10 percent by 0.66 Mev positron emission and 90 percent by K capture and was counted in equilibrium with the 10 minute Cu$^{62}$ daughter which decays 100 percent by emission of a 2.92 Mev positron. The Zn$^{65}$ was counted through 345 mg/cm$^2$ of aluminum absorber which was found to be sufficient to block out all x-rays and particulate radiation, leaving only the 1.11 Mev gamma ray which was assumed to accompany 46 percent of the disintegrations. The counting efficiency of the 1.11 Mev gamma ray was assumed to be 1.11 percent.

Copper Fraction. The activities of Cu$^{61}$, Cu$^{64}$, and Cu$^{67}$ were observed in the copper fractions. The Cu$^{61}$ was counted through 222 mg/cm$^2$ of aluminum absorber which was sufficient to cut out virtually all other activities in the initial portion of the decay. The percentage counting efficiency of the positron of the Cu$^{61}$ through the 222 mg/cm$^2$ of aluminum was determined from the aluminum absorption curve taken for the copper fraction. This percentage was 7.8 percent when the copper sample was on shelf 5 of the counter and the aluminum absorber was on shelf 1 of the counter. These isotopes of copper were assumed to decay as follows: Cu$^{61}$, 66 percent by positron emission;$^{20}$ Cu$^{64}$, 50 percent by $\beta^+$ and $\beta^-$ emission combined; and Cu$^{67}$, 100 percent by $\beta^-$ emission. Air and window absorption correction factors were determined in each case by
straight line extrapolation of aluminum absorption curves. The three isotopes were identified by means of their half-lives and by means of their aluminum absorption curves.

Nickel Fraction. The known activities of Ni\textsuperscript{57}, Ni\textsuperscript{65}, and Ni\textsuperscript{66} were observed in the nickel fractions. In addition to these activities a previously unidentified nickel activity was observed. This activity was identified as Ni\textsuperscript{56}. This Ni\textsuperscript{56} activity was observed in both of the thick targets. Its existence was proved by first assuring that the nickel fraction contained no cobalt at the start of counting. After allowing one of the samples to decay for a period of time, cobalt was milked out and counted. The presence of Ni\textsuperscript{56} was proved by the fact that Co\textsuperscript{56} had grown into the nickel fraction. The Co\textsuperscript{56} was identified by its characteristic aluminum absorption curve and by its half-life. In the two targets where Ni\textsuperscript{56} was observed, the contributions of Ni\textsuperscript{57} and Ni\textsuperscript{66} to the gross nickel decay curve were calculated from their previously determined cross sections and these contributions subtracted from the gross nickel decay curve leaving only the Ni\textsuperscript{56} and Co\textsuperscript{56} components in the curve. Resolution of the Ni\textsuperscript{56} decay curve from this curve indicated that the half-life of the Ni\textsuperscript{56} was approximately six days. The initial slope of the nickel decay curve for a period of four days after milking for cobalt and after all other nickel components had completely decayed indicated a half-life of approximately seven days. The second nickel fraction from a thick target bombardment was allowed to decay without being milked for cobalt.
The decay curve for this sample was identical in form to the decay curve of the sample milked for cobalt up to the time of milking. The complete decay curve eventually tailed out into a 72 day half-life. This portion of the decay was followed through a period of approximately sixty days. Subtraction of this 72 day activity from the gross decay curve left a curve in which the Ni$^{56}$ decay could be seen. The half-life of Ni$^{56}$ here appeared to be approximately 5.5 days. Aluminum and beryllium absorption curves were taken on the Ni$^{56}$ after milking out its Co$^{56}$ daughter. There appear to be three components observable. When counting on shelf two of the counter, the radiation appears to consist of approximately 20 percent gamma rays, approximately 43 percent x-rays from cobalt, and approximately 37 percent of a soft component which is assumed to be particulate radiation. Assuming the counting efficiency of the cobalt x-rays to be 4.5 percent and the counting efficiency of the particle to be 88 percent after correcting for air and window absorption, self-scattering and absorption, and backscattering, one deduces that the total counting efficiency of the Ni$^{56}$ is approximately 10 percent when counted as nickel dimethyl glyoxime of 2.5 mg/cm$^2$ thickness mounted on 1 mil aluminum plate on plexiglass of ~92 mg/cm$^2$ thickness. The counting efficiency of the Ni$^{56}$ in the form of nickel dimethyl glyoxime was also calculated from its relationship with its Co$^{56}$ daughter. In making this calculation it was deduced that the over-all counting efficiency of the Co$^{56}$ was 112 percent under the given conditions. A half-life of six days was assumed for the Ni$^{56}$. Using these figures the counting efficiency of the Ni$^{56}$ was found to be approximately 28 percent. In view of the fact that the absorption curves
were taken on a very low counting sample and that consequently the statistics were none too good, it appears that greater weight should be given to the figures as calculated from the parent daughter relationships (i.e., a counting efficiency of 28 percent). The formation cross sections for the Ni\textsuperscript{56} were calculated from its Co\textsuperscript{56} daughter activity.

One bombardment of elemental iron with alpha particles was made in order to isolate Ni\textsuperscript{56} in larger quantities. The iron was electroplated to a thickness of approximately 85 mg/cm\textsuperscript{2} on a copper probe head which could be water cooled. This target was bombarded with 40 Mev alpha particles in the internal beam of the 60-inch cyclotron. Under these conditions the \( \alpha,2n \) reaction on iron was favored, so that Ni\textsuperscript{56} formed from Fe\textsuperscript{54} was the primary nickel isotope produced. The nickel fraction was separated approximately one week after bombardment in order to allow most of the Ni\textsuperscript{57} produced to decay prior to separation. The nickel fraction was checked for cobalt activity to insure that no cobalt was present in the sample immediately after separation. The sample was placed on a crude beta ray spectrometer immediately after separation. Positively charged particles were present which, without detailed analysis, appeared to consist solely of 0.7 Mev positrons which were probably from Ni\textsuperscript{57}. Negatively charged particles were also present which appeared to be conversion electrons ranging from approximately 0.17 to 0.95 Mev in energy. Approximately 10 days after separation of the nickel fraction it was again examined on the crude beta ray spectrometer. At this time there were again both positive and negative particles present in the radiation. The positively charged particles, without detailed analysis, appeared to
consist solely of 1.5 Mev positrons which were probably from the Co\textsuperscript{56} daughter of Ni\textsuperscript{56}. The negatively charged particles appeared again to be conversion electrons covering a range of energies approximately the same as originally seen.

The nickel fraction was also counted in a scintillation pulse analyzer. Four gamma peaks could be distinguished in this counter. These peaks indicated the presence of gamma rays with energies of approximately 0.14, 0.48, 0.77 and greater than 1.4 Mev respectively. The decay of these peaks was followed on the scintillation pulse analyzer. The 0.14 Mev peak decayed with a half-life of approximately 6 days over a period of about 10 days. The other three peaks tailed off into a longer half-life than this during this time.

For a detailed picture of the decay scheme of Ni\textsuperscript{56} more work and further analysis of the presently available data will be required. It is believed, however, from the work which has already been done that the half-life of Ni\textsuperscript{56} is 6.0 ± 0.5 days. It is also believed that at least a portion of the counting efficiency of the Ni\textsuperscript{56} as originally determined is due to conversion electrons. Whether or not there is any positron decay is at present unclear. It is believed, however, that the positron decay (if any) is certainly less than 24 percent of the total decay, the balance being accounted for by electron capture.

The Ni\textsuperscript{57} was assumed to decay 50 percent by emission of a 845 kev positron and 50 percent by electron capture.\textsuperscript{21} The Ni\textsuperscript{65} was assumed to decay by $\beta^-$ emission as follows: 29 percent, 0.60 Mev; 14 percent, 1.01 Mev; and 57 percent, 2.10 Mev.
The Ni\textsuperscript{66} was counted in equilibrium with its 4.3 minute Cu\textsuperscript{66} daughter. No data were available on the energy of the $\beta^-$ particle emitted by Ni\textsuperscript{66} but from an aluminum absorption curve there appeared to be a component corresponding to a $\beta^-$ particle of approximately 0.34 Mev energy. It was therefore assumed that the Ni\textsuperscript{66} decayed 100 percent by emission of a 340 kev $\beta^-$ particle. Both $\beta^-$ and $\beta^+$ particles were observed on nickel samples placed on the crude beta ray spectrometer.

Cobalt Fraction. The characteristic activities of Co\textsuperscript{55}, Co\textsuperscript{56}, Co\textsuperscript{58}, and Co\textsuperscript{61} were observed in the cobalt fractions. Their decay was assumed to be as follows:

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Percentage Decay</th>
<th>Method of Decay</th>
<th>Energy (Mev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co\textsuperscript{55}</td>
<td>50</td>
<td>positron\textsuperscript{22}</td>
<td>1.01</td>
</tr>
<tr>
<td></td>
<td>50</td>
<td>positron</td>
<td>1.5</td>
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<tr>
<td>Co\textsuperscript{56}</td>
<td>100</td>
<td>positron</td>
<td>1.5</td>
</tr>
<tr>
<td>Co\textsuperscript{58}</td>
<td>14.5</td>
<td>positron</td>
<td>0.47</td>
</tr>
<tr>
<td>Co\textsuperscript{61}</td>
<td>100</td>
<td>$\beta^-$</td>
<td>1.3</td>
</tr>
</tbody>
</table>

The Co\textsuperscript{56} and Co\textsuperscript{58}, having the same half-lives, could not be resolved from the decay curve. The relative abundances of the two isotopes were determined by means of an aluminum absorption curve from which the number of counts due to the 1.5 Mev positron from Co\textsuperscript{56} and the 0.47 Mev positron from Co\textsuperscript{58} could be determined.
In addition to the above mentioned cobalt activities, evidence was observed of the Co\textsuperscript{58m} 9.3 hour activity. The counting efficiency of this activity is quite low and consequently the resolution of the exact decay line is quite difficult. Since the exact counting efficiency is entirely unknown, and resolution of the decay curve is very difficult, no attempt was made to include the cross section for the independent yield of this isomer. The cross section reported for formation of Co\textsuperscript{58} includes the independent yield of Co\textsuperscript{58m}.

**Iron Fraction.** The characteristic activities of Fe\textsuperscript{52} and Fe\textsuperscript{59} were observed in the iron fractions. The presence of $\beta^+$ particles emitted from the iron fraction approximately 8 hours after bombardment indicated the presence of Fe\textsuperscript{52}, a fact which was confirmed by the decay curve. The presence of a $\beta^-$ particle at the same time indicated the presence of Fe\textsuperscript{59} or a higher isotope. The relative abundance of the $\beta^-$ particle was quite low and did not seem to indicate the presence of any higher isotope than Fe\textsuperscript{59}, nor was the presence of any other isotopes evident in the decay curves.

The Fe\textsuperscript{52} was counted in equilibrium with its 21 minute Mn\textsuperscript{52} daughter. It was assumed to decay 38.5 percent by emission of a 640 kev positron and 61.5 percent by electron capture.\textsuperscript{23} The characteristic energies of the 2.66 Mev positron from Mn\textsuperscript{52} daughter and the 0.640 Mev positron from Fe\textsuperscript{52} were evident in aluminum absorption curves taken on the iron fraction.

The Fe\textsuperscript{59} was assumed to decay 100 percent by emission of 0.46 Mev $\beta^-$ particles. The 260 kev $\beta^-$ previously reported for this isotope was
not resolvable from any of the aluminum absorption curves taken on the Fe\(^{59}\). The absorption curves taken were resolved into a gamma and 460 kev \(\beta^-\) component. The air and window absorption corrections were therefore made on the basis of the absorption curves actually recorded.

**Manganese Fraction.** The characteristic activities of Mn\(^{51}\), the 5.8 day Mn\(^{52}\), Mn\(^{54}\), and Mn\(^{56}\) were identified in bombardments number 3 and 4. Their decay was assumed to be as follows:

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Percentage Decay</th>
<th>Method of Decay</th>
<th>Energy (Mev)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mn(^{51})</td>
<td>100</td>
<td>positron</td>
<td>2.4</td>
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<td>65</td>
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<td></td>
</tr>
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<td>EC</td>
<td></td>
</tr>
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<td>Mn(^{56})</td>
<td>15</td>
<td>(\beta^-)</td>
<td>0.73</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>(\beta^-)</td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>60</td>
<td>(\beta^-)</td>
<td>2.86</td>
</tr>
</tbody>
</table>

The manganese fraction was separated too late in bombardment number 5 for the 44.5 minute Mn\(^{51}\) to be identified. In the two "thick" targets (bombardments number 11 and 12) the manganese fractions were not separated early enough for anything but the Mn\(^{52}\) and Mn\(^{54}\) activities to be identified. The isotopes were identified by their half-lives, by their absorption curves through aluminum, beryllium and lead absorbers and by the sign of the particulate radiation.
Due to the long half-life of the Mn$^{54}$ it was necessary to resolve its decay curve analytically rather than follow the decay through several half-lives as would have been preferable were the time available.

The manganese results obtained are probably the least accurate of any of the fractions due to the great difficulty in obtaining a pure sample and due to the complexity of the decay curves. The manganese dioxide precipitate has a great tendency to carry other activities with it. A pure manganese decay curve from a fraction isolated soon enough to observe the 44.5 minute Mn$^{51}$ will contain this 44.5 minute activity, a 26 day activity from its Cr$^{51}$ daughter, the 2.6 hour activity of Mn$^{56}$, the 5.8 day activity of Mn$^{52}$ and the 310 day activity of Mn$^{54}$.

**Chromium Fraction.** The characteristic activities of Cr$^{48}$, Cr$^{49}$, and Cr$^{51}$ were observed in all the chromium fractions separated from "thin" targets. The chromium fractions from the two thick targets were separated too long after bombardment for Cr$^{48}$ and Cr$^{49}$ to be observed, and Cr$^{51}$ was the only chromium activity observed in these two fractions.

The Cr$^{48}$ previously reported by Rudstam was very difficult to resolve from the decay curves due to its low counting efficiency. Its presence was definitely indicated, however, by the presence of the 16 day V$^{48}$ daughter activity in the decay curves. Cross sections for this isotope were calculated from the observed activity of its 16 day V$^{48}$ daughter.

The Cr$^{49}$ appeared to have a half-life of 42.5 minutes. This isotope was assumed to decay 100 percent by the emission of a 1.45 Mev positron.
The Cr\textsuperscript{51} appeared to have a half-life of 26.5 days. The over-all counting efficiency of this isotope was assumed to be 2.6 percent in accordance with the determination made by Batzel.\textsuperscript{7}

**Vanadium Fraction.** The characteristic activity of V\textsuperscript{47} was observed in one vanadium fraction along with the characteristic activity of V\textsuperscript{48}. The latter activity was also observed in a second vanadium fraction which was separated too late for the V\textsuperscript{47} to be observed. These activities were recognized by the decay curves and aluminum absorption curves. The V\textsuperscript{47} was assumed to decay 100 percent by positron emission. Air and window absorption correction was determined from the aluminum absorption curve taken for this activity. The V\textsuperscript{48} was assumed to decay 58 percent by positron emission and 42 percent by electron capture. Air and window absorption correction was determined from the aluminum absorption curve taken for this activity.

**Titanium Fraction.** Ti\textsuperscript{45} was the only activity observed in the titanium fractions. This isotope was assumed to decay 100 percent by the emission of a 1.0 Mev positron.

**Calcium Fraction.** The 152 day Ca\textsuperscript{45} and the Ca\textsuperscript{47} activities were the only two observed in the calcium fractions. The Ca\textsuperscript{45} was assumed to decay 100 percent by the emission of a 0.25 Mev beta particle. The calcium decay curves were identical with those reported by Batzel\textsuperscript{7} in his work with spallation products of copper. The half-life of Ca\textsuperscript{47} was
taken as 4.9 days. The Ca\(^{47}\) was assumed to decay 100 percent by the emission of a 1.1 Mev beta particle and was counted in equilibrium with its 3.4 day Sc\(^{47}\) daughter which was assumed to decay 100 percent by the emission of a 0.61 Mev beta particle. The two calcium isotopes were identified by their half-lives and by their aluminum absorption curves.

**Potassium Fraction.** Two potassium activities were observed in the potassium fractions isolated, K\(^{42}\) and K\(^{43}\). The K\(^{42}\) was assumed to decay 75 percent by emission of a 3.58 Mev beta particle and 25 percent by emission of a 2.04 Mev beta particle. The K\(^{43}\) was assumed to decay 50 percent by emission of a 0.24 Mev beta particle and 50 percent by emission of a 0.81 Mev beta particle.

**Sulfur Fraction.** S\(^{35}\) was the only activity observed in the sulfur fractions isolated. This isotope was assumed to decay 100 percent by emission of a 0.167 Mev beta particle and was identified by means of its characteristic 87 day half-life.

**Phosphorus Fraction.** The only activity observed in the phosphorus fractions isolated was that of the 14 day P\(^{32}\) which was identified by its half-life and aluminum absorption curve. A slight tailing into a longer activity than the 14 day P\(^{32}\) was observed in all the phosphorus fractions. The longer lived activity may perhaps be the 25 day P\(^{33}\) which has recently been reported.\(^{24}\) Insufficient time, however, was available for the half-life of this activity to be established definitely. The P\(^{32}\)
was assumed to decay 100 percent by emission of a 1.71 Mev beta particle.

Sodium Fraction. The activity of $\text{Na}^{24}$ was observed in the sodium fractions. Evidence of the presence of $\text{Na}^{22}$ was observed in the tailing off of the decay curve. The samples were counted for an insufficient length of time, however, to identify definitely the long lived activity. The $\text{Na}^{24}$ was assumed to decay 100 percent by emission of a 1.39 Mev beta particle followed by emission of a 2.76 Mev and a 1.38 Mev gamma ray in cascade.

B. Summary of Cross Sections Determined

In the table below are listed the cross sections (in millibarns) for formation of the various isotopes identified. Seventeen separate bombardments were made during the course of this work. The cross sections for formation are listed under the particular bombardment from which they were determined. The shortest lived activity identified was the 33 minute $\text{V}^{47}$. The longest lived activity identified was the 310 day $\text{Mn}^{54}$. In the latter case and in the case of the 250 day $\text{Zn}^{65}$ and the 152 day $\text{Ca}^{45}$ it was impossible due to lack of time to follow the decay through as much as one half-life. There was, however, no apparent deviation of decay curves from the proper half-life during the time that the decay was followed. The decay of $\text{S}^{35}$ was followed through a little more than one half-life. The decay of all other isotopes reported was followed through at least two half-lives and in most cases through considerably more than this (see table of cross sections).
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<th>2</th>
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<th>4</th>
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Were more time available, the cross sections for several other radioactive isotopes could undoubtedly be obtained. The ones listed above, however, are sufficiently widely distributed that they give a very good indication of the cross sections for all the isotopes. Illustrative of this fact is Figure 4 in which the cross sections determined are plotted on a chart of Z versus N. The cross sections having the same order of magnitude are enclosed within "isobarns." It is readily apparent from this chart that all the cross sections from 10 to 99 mb fall on one continuous area on the chart, those from 1 to 9.9 mb fall in another area surrounding the first, etc. It is believed that from such a chart the yields of isotopes not directly determined (including stable isotopes) can be estimated with a fair degree of accuracy.

The cross sections reported under bombardments number 1, 2, 3, 7, 8, 9, 10, 13, 14, and 15 were calculated relative to the cross section for formation of Na\(^{24}\) from aluminum (10 mb), all of the targets in these bombardments having been "thin" targets. Difficulties were encountered in cutting the aluminum monitor foils and zinc target foil evenly in the "thin" targets of bombardments number 4, 5, and 6. The cross sections reported under these three bombardments, therefore, were calculated relative to the cross section for formation of Cu\(^{67}\) from zinc (4.7 mb). Bombardments number 11 and 12 were "thick" targets. The cross sections

*The term "isobarn" has been coined to describe a line connecting nuclides having equal cross sections for formation."
reported for these bombardments were calculated relative to the cross section for formation of Cr\textsuperscript{51} from zinc (15 mb). The cross sections reported under bombardments number 16 and 17 were calculated relative to the cross section for formation of Cu\textsuperscript{61} from zinc (33 mb).

It will be noted that the cross section for Cu\textsuperscript{67} in bombardments number 1, 7, and 8 check quite well, while those in bombardments number 3, 11, and 12 do not correspond to the previously mentioned bombardments. It is believed that the cross sections for all three copper isotopes in bombardment number 3 are in error. This bombardment was virtually identical with numbers 1, 7, and 8 except that the chemical recovery of copper in bombardment number 3 was quite low. For this reason the results for copper in bombardment number 3 are disregarded. The results for Cu\textsuperscript{67} in bombardments number 11 and 12 are not so easily explained. It was originally planned to use Cu\textsuperscript{67} as internal monitor on the thick targets, inasmuch as its decay characteristics, chemical separation procedure, etc. made it a convenient isotope with which to work. When the cross sections in bombardments number 11 and 12 were calculated relative to Cu\textsuperscript{67}, however, it was discovered that they did not check with those cross sections determined in thin targets. The variation between the two types of targets was markedly similar in those cases where a good cross check was possible. The energy of a 340 Mev proton beam passing through 3/4 of an inch of zinc will be decreased only on the order of 10 percent. Hence it seemed unlikely that cross sections should vary much due to the decrease in energy of the beam. It seems more reasonable to assume that the apparent cross section of Cu\textsuperscript{67} varies with the
thickness of the target. This isotope can be formed in only a limited number of different ways from the zinc nuclei. One likely method of formation is through the action of secondary neutrons on $\text{Zn}^{68}$, $(\text{Zn}^{68}(n,\text{pn})\text{Cu}^{67})$. In thick targets, this reaction would probably be much more pronounced than in a thin target resulting in an increase in the cross section for formation of $\text{Cu}^{67}$. It is therefore believed that the apparent cross section for formation of $\text{Cu}^{67}$ is actually some 45 to 50 percent higher in a target 3/4 inches thick than it is in a 5 mil zinc target. The cross sections of a few other of the observed isotopes might be affected in a like manner but probably not to as great an extent as that of $\text{Cu}^{67}$. It was because of this apparent change in the cross section of $\text{Cu}^{67}$ with thickness of target that $\text{Cr}^{51}$ was used as a monitor in the thick targets of bombardments number 11 and 12. It is believed to be purely a coincidence that the cross section of $\text{Cu}^{67}$ in bombardment number 3 is similar to those in bombardments number 11 and 12.

It will be noted that the cross sections for $\text{Fe}^{52}$ and $\text{Fe}^{59}$ in bombardment number 3 do not correspond with those in bombardments number 1 and 6. The ratios between the cross sections for the two isotopes are comparable in all three cases, however. A slight amount of inactive impurity in the final weighing of the iron sample would decrease the calculated cross section by an appreciable amount as would a simple error in the weighing. It is believed that something like this occurred in bombardment number 3 to account for the discrepancy in the iron results. The results for iron in bombardment number 3 are, therefore, disregarded in averaging the cross sections.
The reasons for the cross section of Co\textsuperscript{58} being higher in bombardment number 5 than in the two other bombardments is unknown. The result in bombardment number 5 is disregarded, and the cross section for formation of Co\textsuperscript{58} is taken as the average of the cross sections determined in bombardments number 2 and 3.

The manganese decay curves were quite difficult to resolve. In the two short bombardments where manganese was separated early enough to observe the 44.3 minute Mn\textsuperscript{51}, the resolution of the decay curve was complicated by the 26.5 day Co\textsuperscript{51} activity which grew in from the Mn\textsuperscript{51}. It was hoped that a good check on the cross sections of Mn\textsuperscript{52} and Mn\textsuperscript{54} could be obtained from the two thick target bombardments. The decay of these two samples, however, had not progressed sufficiently for the two activities to be resolved prior to the completion of this work. There appeared to be present some other activity than the 5.8 day Mn\textsuperscript{52} and the 310 day Mn\textsuperscript{54}. An analysis of the manganese fraction on the crude beta ray spectrometer approximately 15 days after the end of bombardment showed beta particles present in higher abundance than would be expected from Mn\textsuperscript{54}. Indeed one would not expect to detect the $\beta^-$ particles from this source at all under the given conditions. Whether these $\beta^-$ particles were from an impurity in the manganese fraction or whether they were from a previously unreported isotope of manganese (perhaps Mn\textsuperscript{57}) could not be determined. Additional work on this problem would probably be worthwhile in an attempt to identify a new isotope of manganese. The fact that some impurity (probably cobalt) was present in the thick targets was proved by the presence of positrons 87 days after bombardment.
Beta particles were also present at this time. One of the thick samples was repurified approximately 90 days after bombardment with a subsequent loss in activity of approximately 50 percent which verified the presence of impurities.

The cross section for Mn$^{54}$ reported under bombardment number 12 was calculated on the assumption that all of the activity after repurification consisted of Mn$^{54}$. This assumption was probably incorrect as evidenced by the fact that the cross section as calculated is considerably higher than that calculated for the other three bombardments.

The cross sections for Mn$^{52}$ were calculated from bombardments number 11 and 12 by assuming that virtually all of the counts immediately after separation of the manganese fractions (approximately 10 days after bombardment) were due to the 5.8 day Mn$^{52}$. Assuming that at least one other activity in addition to Mn$^{54}$ was also present would account for the fact that these cross sections are higher than those calculated from bombardments number 3, 4, and 5. The results for bombardments 11 and 12 are disregarded in computing the average cross sections for manganese isotopes.

The results for chromium isotopes calculated from bombardment number 6 are far different from those calculated from bombardments number 8 and 9. The chemistry for chromium was quite difficult and considerable trial and error was necessary before a good procedure was finally settled upon. The physical appearance of barium chromate from bombardment number 6 indicated that something was wrong with the sample. Possibly the chemical purification was poor although this was not evident in the decay
curves. The results for chromium from bombardment number 6 are therefore disregarded in averaging the results for chromium isotopes.

The counts on $^{85}$S from the "thick" target in bombardment number 7 were only slightly above background due to its low cross section and low counting efficiency. It is therefore believed that the cross section determined from bombardment number 12 is probably closer to the true value than is the cross section determined from bombardment number 7.

Aluminum monitors were used with the "thin" target of bombardment number 15. Inasmuch as sodium was removed from this target, this was an error which unfortunately was not realized until too late. It resulted in the cross section of Na$^{24}$ appearing higher than it should have due to the fact that some Na$^{24}$ nuclei would recoil from the aluminum foils and lodge in the zinc foil. The cross section for formation of Na$^{24}$ is taken as the average of the results obtained in bombardments number 16 and 17.

IV. CONCLUSIONS

If we conceive of a nucleus as a sphere containing an assemblage of protons and neutrons, we can define the relative probability of a certain reaction occurring between this nucleus and an incident particle, such as a proton, in terms of the cross section of the target nucleus. If the incident particle is charged, it must have sufficient energy to penetrate the coulombic potential barrier between itself and the nucleus before there is much chance of any reaction occurring. The cross section generally rises slowly with increasing energy of the incident particle
and then falls off as other more complicated processes become more likely with increasing energy. From these and other facts the theories of the "compound nucleus" and the "liquid drop model" were developed in which the incident particle strikes the nucleus, is absorbed in it to form a "compound nucleus," and then certain particles, depending mainly upon the nature of the target and energy of the incident particle, "boil off" as from a liquid drop.

These theories, however, do not hold for high energy reactions. From these theories we would expect that the higher the energy of the incident particle, the greater the number or the more complex would be the particles boiled off. Such is clearly not the case as is illustrated by the results of the present work as well as other work on spallation. There is a much greater probability of forming isotopes in the immediate neighborhood of the target nucleus (i.e., where few particles are boiled off) than far removed from the target nucleus (i.e., where many particles are boiled off).

This apparent anomaly may be explained by Serber's theory of nuclear transparency. According to this theory the mean free path of a particle traversing nuclear matter increases with its energy, and at very high energies the nucleus begins to be transparent or partially transparent to the bombarding particle. He estimates the mean free path in nuclear matter of a 100 Mev nucleon to be about 4 x 10^{-13} centimeters with an average transfer of energy of about 25 Mev per collision. Batzel in his work on copper has estimated for a 240 Mev proton a mean free path of about 4 x 10^{-13} cm and a mean energy loss per collision of about 70 Mev.
These figures for copper should also hold for zinc since the two elements are adjacent in the periodic chart. The 34.0 Mev proton "striking" a zinc nucleus is then much more aptly described as "passing through" the zinc nucleus. In this process there is much greater likelihood of its suffering only a few collisions with individual nucleons within the nucleus, transferring only a part of its energy to each nucleon struck, and passing on out of the nucleus with a considerable amount of its original energy, than there is of its being captured by the nucleus and forming a high energy "compound nucleus" from which various particles boil off. An individual nucleon within the nucleus having been struck by the incident proton may itself be knocked out of the nucleus without sharing any of its energy with the other nucleons within the nucleus. On the other hand the struck nucleon may in turn strike other nucleons within the nucleus and eventually share part or all of its energy with the nucleus as a whole, forming a "compound nucleus" or what we might more correctly describe as a "secondary compound nucleus." The most likely particles to boil off from a compound nucleus would appear to be neutrons since they would not have the problem of penetrating the coulombic potential barrier. The high cross sections for the zinc isotopes demonstrate the high probability of non-capture reactions involving the emission of neutrons only. Nuclei excited to a greater degree will have the emission of charged particles such as protons, deuterons, and alpha particles begin to compete with the emission of neutrons. This fact is illustrated by the similarity of cross sections along 45 degree lines through the target nuclei on a Z versus N plot.
Gallium isotopes are in all probability formed by the incident protons striking a neutron, transferring most or all of its energy to the neutron and knocking it out of the nucleus. One neutron or more might then boil off the compound nucleus depending upon whether or not it was excited and if so to what extent. A part of the formation might be accounted for by the reaction of secondary protons with zinc nuclei. Protons knocked out or boiled off one zinc nucleus with much lower energy than the original beam of 340 Mev protons might in turn strike other zinc nuclei, be captured by them, and give rise to the formation of gallium isotopes through $(p,xn)$ or $(p,\gamma)$ reactions. The probability of these proton capture processes at high energy is obviously low in relation to noncapture processes, however.

In order to yield isotopes far removed from zinc by spallation reactions, the compound nucleus would have to be highly excited. The fact that the cross sections fall off as the isotopes are further and further removed from the target nucleus is indicative of the fact that in only a relatively few collisions does the incident particle transfer all (or nearly all) of its energy to the target nucleus.

Most of the cross sections determined for the various isotopes are partially chain yields of their isobars, due to the finite time of bombardment and time required for separation of the elements. It will be noted, however, that Cu$^{64}$, Co$^{58}$, and Mn$^{54}$ are shielded nuclides. Their formation could not be accounted for by a beta or positron chain decay. These nuclides must have been formed directly from the target nuclei by the boiling or splitting off of small nuclear fragments. Their cross
sections being quite high leads one to assume that the greater portion of each cross section reported is independent of beta chain decay.

A small portion of each cross section is probably accounted for by the emission of particles larger than alpha particles from the excited nuclei. Evidence of this fact has previously been reported by Batzel\textsuperscript{29} in his work with medium weight elements. As he states, such a process would more correctly be described as "fission" than as "spallation." Wright\textsuperscript{30,31} has found evidence of the emission of Li\textsuperscript{8} particles as such and Marquez and Perlman\textsuperscript{32} have found evidence of the emission of Be\textsuperscript{7} from excited nuclei.

**SUMMARY**

From the work which has been done it may be inferred that with high energy proton bombardment of zinc the following, in broad outline at least, is a picture of what happens:

(1) When the nucleus is struck a great variety of processes are possible ranging from the incident particles passing through most of the nucleus without any reaction and suffering perhaps one collision without transferring any appreciable amount of its energy to the nucleus, to the capture of the incident particle with all or practically all of its energy by the nucleus and the subsequent boiling off of small particles or fission into relatively large particles.

(2) Probability of capture of the incident proton without boiling off any particles other than neutrons is relatively low.
(3) Of much higher probability is the transfer of a part of the proton's energy to the nucleus without capture of the proton, followed by boiling off of neutrons only.

(4) Of high probability also is the transfer of a still greater portion of the proton's energy to the nucleus and the subsequent boiling off of charged particles to give isotopes of elements several Z below the target nuclei. The number of protons and neutrons emitted is roughly equivalent as indicated by the similarity of cross sections on 45 degree lines through the target nuclei on a Z versus N plot.

(5) The relative yield in general of isotopes fall off the further removed they are from the target element in the periodic table.

(6) The relative yield in general of the isotopes of any element fall off the further removed the nuclide is from the line of stability. Or in other words a line connecting the isotopes of highest yield of all the elements considered will follow roughly the line of stability except that in its upper end it will tend to bear toward the neutron deficient side of stability.
V. ACKNOWLEDGMENTS

The cooperation and assistance of numerous people has made this work possible. Without this cooperation and assistance the completion of this work would have been impossible. To all these persons, too numerous to list in detail, I wish to express my gratitude and thanks.

I particularly wish to thank Dr. Glenn T. Seaborg under whose direction and guidance this work was performed. I also wish to thank Dr. Peter C. Stevenson, Dr. Robert L. Folger, Dr. Harry G. Hicks and Mr. Walter E. Nervik for their assistance and advice.

The bombardment of iron with alpha particles and the subsequent work on Ni\textsuperscript{56} from this bombardment was performed in cooperation with Mr. Per Kofstad.

The cooperation of Mr. James T. Vale and the group operating the 184-inch cyclotron is much appreciated and gratefully acknowledged.

This work was performed under the auspices of the United States Atomic Energy Commission.
VI. BIBLIOGRAPHY

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VII. LIST OF ILLUSTRATIONS

Figure 1. Typical "Thin" Target.
Figure 2. Typical "Thick" Target.
Figure 3. Typical Mountings Used in Counting Samples.
Figure 4. Isobarn Plot of Cross Sections.
Figure 5. Experimentally Determined Cross Sections on a Z versus N Plot of the Isotopes.
Fig. 1

EXPLODED VIEW OF
TARGET FOILS

1 mil ALUMINUM
GUARD FOILS

1/2 mil ALUMINUM
MONITOR FOILS

5 mil ZINC FOIL

TARGET FOILS

340 MEV
PROTON BEAM

SCREW TO CLAMP
TARGET FOILS IN
HOLDER

COPPER TARGET
HOLDER

FIG. 1

appx. 1/4" 
APPX 3/8"
SEVERAL 5mil ZINC FOILS WRAPPED IN 1mil ALUMINUM ENVELOPE

340 MEV PROTON BEAM

FIG. 2
FIG. 3

4 sq. cm. SAMPLE DISH OF 1 mil ALUMINUM

SCOTCH TAPE

ALUMINUM FRAME

CARD BOARD

NOTE: THIS ARRANGEMENT ALSO USED FOR COUNTING MONITOR FOILS

A

THICK ALUMINUM PLATE

SAMPLE

B

PLEXIGLASS

4 sq. cm. SAMPLE DISH OF 1 mil ALUMINUM

ALUMINUM HOLDER RECESSED TO HOLD PLEXIGLASS PLATE

C

1 mil ALUMINUM DISH WITH 1 cm² DEPRESSION

MU 3097

Fig. 3
Fig. 4

Curve I encloses isotopes with cross sections from 1 to 99 millibarns
Curve II from 1 to 9.9 millibarns
Curve III: 0.1 to 0.99
Curve IV: 0.01 to 0.099
Curve V: 0.001 to 0.0099

- Target nuclei
- Stable isotopes
- Cross sections determined experimentally

Fig. 4
FIGURES DENOTE EXPERIMENTALLY DETERMINED CROSS SECTIONS IN MILLIBARNS

Fig. 5