# Nuclear Reactions of Iron with 340-Mev Protons\*

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Natural iron has been bombarded with the full energy proton beam of the Berkeley 184-inch synchrocyclotron, and a number of radioactive spallation products have been isolated and identified and their formation cross sections measured against that of the reaction  $Al^{27}(p, 3pn)Na^{24}$ . A simple mechanism has been postulated to account for the observed product yields. A new isotope of chromium, assigned to mass 48, has been observed and partially characterized.

## I. INTRODUCTION

A SERIES of bombardments of iron with 340-Mev protons has been carried out in the circulating beam of the Berkeley 184-inch cyclotron with the purpose of studying the cross section for formation of the various spallation or fission products. It has been found possible to isolate a number of radioactive product nuclides by standard radiochemical techniques using inert carriers and to identify the nuclides so obtained by observing their half-lives and radiation characteristics. Both absorption measurements and low resolution magnetic beta-spectrometer measurements were used where appropriate to differentiate between isotopes of the same element with similar half-lives.

A mechanism of spallation has been postulated which contains the basic assumption that the probability of emitting a proton from a highly excited nucleus of low atomic number (i.e., less than 30) is approximately equal to the probability of emitting a neutron from the same nucleus under the same conditions. From the postulated mechanism it can be deduced that the spallation products for a given nuclide can be divided into groups *a priori*, the yields of the products in any given group falling on a smooth curve when plotted against mass number. Five such groups were predicted for the spallation products of iron, considering only the abundant isotope Fe<sup>56</sup>. Experimental results show satisfactory agreement with the hypothesis.

#### **II. EXPERIMENTAL TECHNIQUES**

# A. Target Makeup

Two types of targets were used which may be referred to for convenience as "thin" and "thick." Each thin target consisted of a single iron foil approximately ten one-thousandths of an inch in thickness through which the beam passed perpendicular to the plane of the foil. On either side of the target foil were "guard foils" of 0.001-inch thick aluminum, and outside and parallel to these were foils of 0.0005-inch thick aluminum (see Fig. 1) used to monitor the total beam through the target by use of the  $Al^{27}(p, 3pn)Na^{24}$  reaction as described by Stevenson and Folger.<sup>1</sup> Energy degradation of a 340-Mev proton beam in passing through such a foil is negligible.

Each thick target consisted of a stack of from four to six 0.01-inch iron foils, the dimensions of the stack being about  $1\frac{3}{4} \times \frac{1}{2}$  inches, the beam passing through the stack parallel to the  $\frac{1}{2}$ -inch dimension. The energy degradation undergone by a 340-Mev proton beam in  $\frac{1}{2}$  inch of iron is calculated to be about 20 Mev.<sup>2</sup> The beam in this case was monitored by observing the yield of Fe<sup>52</sup>, previously calibrated against the Al<sup>27</sup>(p, 3pn)Na<sup>24</sup> reaction in thin target bombardments.

The iron used was of high purity. A spectrographic analysis performed by Conway and Moore of this laboratory showed very slight amounts of a small number of impurities. These impurities are tabulated in Table I.

Seven bombardments were made. Table II lists the data pertinent to each bombardment.

### **B.** Chemical Separations

Usually 10 mg of each element to be isolated was added as carrier to the dissolving medium prior to addition of the target. The chemical methods used in the bombardments consisted of specific or group precipitations or solvent extractions for each element sought, combined with precipitations of unwanted elements (added as carriers) by reagents not precipitating the desired elements (so-called "scavenge" precipitations). The precise combinations of extractions, specific precipitations, and scavenges used varied depending on which combinations of elements were to be isolated. However, the more important steps in the isolation and purification of the different elements are given below. The percentage recovery of the activities formed was determined gravimetrically by weighing the recovered carrier, complete exchange being assumed. The target was dissolved in approximately 12 N HCl (bombardments FeP1 to FeP5) or in approximately 3 N HNO<sub>3</sub> (bombardments *FeP*6 and *FeP*7).

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<sup>&</sup>lt;sup>1</sup> P. C. Stevenson and R. L. Folger (unpublished work).

<sup>&</sup>lt;sup>2</sup> Aron, Hoffman, and Williams, AEC Unclassified Document No. 663 (May 28, 1951).

# 1. Cobalt

Cobalt was precipitated from ammoniacal solution with hydrogen sulfide gas. The sulfide was dissolved in hydrochloric acid containing potassium chlorate, and cobalt was then precipitated by the use of potassium hydroxide. This precipitate was dissolved in acetic acid, and potassium nitrite was added to precipitate  $K_3Co(NO_2)_6 \cdot H_2O$ . This precipitate was washed, dried at 110°C on an aluminum dish, and weighed. In bombardments *FeP1* and *FeP2* an extraction of cobaltthiocyanate complex into ethyl acetate was used as an additional purification step.

### 2. Iron

The principal step in the iron isolation was the extraction of ferric chloride from 7.5 N HCl into isopropyl ether. The extraction is specific for iron in this region of the periodic table. The ether layer was washed three times with 7.5 N HCl, then iron was re-extracted into water and ferric hydroxide precipitated by the addition of ammonium hydroxide. This precipitate was dissolved in 7.5 N HCl and the cycle repeated. The final ferric hydroxide precipitate was washed, dried, and ignited on a platinum dish to Fe<sub>2</sub>O<sub>3</sub> and weighed. In bombard-

TABLE I. Spectrographic analysis of target iron.

Impurity	Amount, parts per million by weight
As	Not detected (<100)
Co	60
Mn	Not detected $(<100)$
Ni	150
Cu	<50

ment FeP2 ferric-thiocyanate complex was also extracted into ethyl acetate and re-extracted into aqueous ammonia. In bombardments FeP1 and FeP2 iron was precipitated as ferric phosphate from dilute hydrochloric acid, washed, dried, and weighed as  $FePO_4 \cdot H_2O$ .

#### 3. Manganese

Manganese was precipitated as manganese dioxide by potassium chlorate from a fuming nitric acid solution. The precipitate was dissolved in hydrochloric acid containing hydrogen peroxide. Tartaric acid and small amounts of chromium, titanium, and vanadium were added and manganous sulfide was then precipitated from alkaline solution. The sulfide was dissolved in concentrated nitric acid and manganese dioxide was brought down from fuming nitric acid solution as before. The whole cycle was repeated once. The  $MnO_2$ precipitate was then dried at  $110^{\circ}C$  and weighed.

### 4. Chromium

In bombardment FeP3 chromium was separated as the iodate from 1 N HCl (after removing titanium and iron). The iodate was destroyed by reduction with sulfur dioxide, and chromium was precipitated as the

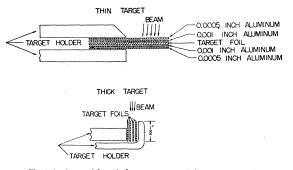


FIG. 1. Assembly of the two types of targets used.

hydroxide. The hydroxide was oxidized to chromate with basic peroxide solution, and the chromium was extracted as perchromic acid from dilute perchloric acid into amyl acetate. The amyl acetate layer was washed and chromium was re-extracted into aqueous ammonia. The perchromic acid extraction was repeated. Finally, chromium was precipitated from acetic acid solution as  $BaCrO_4$ , washed, dried at  $110^{\circ}C$ , and weighed.

In bombardment FeP5 chromium was first brought down as barium chromate, then extracted into amyl acetate from a perchloric acid solution containing hydrogen peroxide. In bombardment FeP2 chromium was oxidized with potassium bromate in perchloric acid and then extracted into ether after cooling and addition of hydrogen peroxide.

### 5. Vanadium

Vanadium was separated as sulfide from ammoniacal solution. The sulfide was dissolved in concentrated nitric acid, scavenged with manganese dioxide, precipitated with potassium chlorate and fuming nitric acid and then with yttrium hydroxide in ammoniacal solution. Vanadium sulfide was again precipitated. The sulfide was dissolved and the solution was buffered with sodium acetate-acetic acid. Chromium was then added to act as holdback and vanadium was oxidized by hydrogen peroxide. Lead vanadate was precipitated. The precipitate was dissolved in 2 N HNO<sub>3</sub>, and lead was removed as the sulfide. The solution was then buffered with sodium acetate-acetic acid and oxidized and Pb(VO<sub>3</sub>)<sub>2</sub> was again brought down. This was washed, dried at 100°C, and weighed.

TABLE II. Summary of bombardments.

Bombard- ment No.	Duration of bombardment (min)	Type of target	Elements isolated
FeP1	20	Thin	Fe, Co
FeP2	46	Thin	Fe, Co, Cr
FeP3	46	Thin	Fe, Co, Cr, Mn, V
FeP4	17	Thick	Fe, Mn, Ca, Ti, Sc
FeP5	109	Thick	Fe, Co, Cr, Mn, Na, S, P, Ca
FeP6	65	Thin	Fe, V, Sc, K, Si, Cl
FeP7	55	Thin	Fe, Ti, K, Cl, Si, Cr

# 6. Titanium

For the separation of titanium the precipitation of titanium iodate from 6 N HNO<sub>3</sub> was used. The precipitate was dissolved in hydrochloric acid containing sodium sulfite. Titanium was then precipitated as hydroxide with ammonia. The hydroxide was dissolved in 0.5 N HNO<sub>3</sub> and scavenged with yttrium oxalate. Tartaric acid was added and the solution made ammoniacal to be scavenged with iron and manganese sulfides. The supernatant from the last scavenge was made 1 Nin hydrochloric acid, and titanium was precipitated with cupferron. The precipitate was then metathesized to  $TiO_2 \cdot H_2O$  with ammonium hydroxide. The hydrous oxide was dissolved in hydrochloric acid and the iodate precipitated. The iodate was dissolved in hydrochloric acid and sodium sulfite. Finally, titanium was precipitated as hydroxide with ammonia, washed, ignited to  $TiO_2$ , and weighed.

### 7. Scandium

Scandium was separated by precipitation with oxalic acid from 0.5 N HNO<sub>3</sub>. The precipitate was dissolved in concentrated nitric acid; manganese carrier and fuming nitric acid were then added. Potassium chlorate was added to precipitate manganese dioxide and to destroy the oxalate. Scandium was precipitated as the hydroxide by adding ammonia gas to the supernatant. After two more oxalate-hydroxide cycles the scandium hydroxide was washed, ignited to  $Sc_2O_3$  on a platinum dish, and weighed.

# 8. Calcium

Calcium was precipitated as calcium oxalate with oxalic acid (bombardment FeP4) or as calcium carbonate from alkaline solution. The calcium precipitates were dissolved in nitric acid (plus potassium chlorate in bombardment FeP4). The solution was made alkaline with ammonium hydroxide and scavenged with yttrium and titanium hydroxides, then with manganese dioxide (precipitated by hydrogen peroxide), and finally with cobalt sulfide. The supernatant was acidified with acetic acid, and calcium was precipitated with oxalic acid. The cycle was repeated twice. Finally, the calcium oxalate precipitate was washed, dried at 110°C, and weighed as CaC<sub>2</sub>O<sub>4</sub>·H<sub>2</sub>O.

### 9. Potassium

After the precipitation of most elements with ammonium hydroxide, potassium was left in solution. This supernatant was made about 0.1 N in acid, phosphate ion added, and then ferric ion in excess to bring down ferric phosphate. The solution was then made alkaline with ammonium hydroxide and scavenged twice with iron and titanium hydroxides, then twice with chromium sulfide and with calcium carbonate or calcium oxalate. The solution was then acidified with hydrochloric acid and evaporated to dryness in a porcelain casserole. The ammonium salts were removed by fuming over a Meker burner. The residue was dissolved in water. Perchloric acid was added, the solution was heated to fuming, then cooled in an icebath, and absolute alcohol was added to precipitate potassium perchlorate. This was carefully washed with absolute alcohol to remove ammonium perchlorate and recrystallized twice from perchloric acid. The precipitate was washed with alcohol, dried at 110°C, and weighed as KClO<sub>4</sub>.

## 10. Chlorine

When chlorine was to be isolated, the carrier was added as solid, freshly prepared silver chloride, and the target was dissolved in dilute ( $\sim 3 N$ ) nitric acid. The exchange between chloride ion in the silver chloride precipitate and the radioactive chloride ion in the solution is supposed to be rapid and complete.<sup>3</sup> After the target had completely dissolved, the silver chloride precipitate was separated. Silver chloride was then dissolved in 5 N NH<sub>4</sub>OH and scavenged with ferric hydroxide. Potassium iodide was then added to precipitate silver nitrate was added to remove iodide. The supernatant was acidified with nitric acid to precipitate silver chloride and the precipitate was washed, dried at 110°C, and weighed.

#### 11. Sulfur

The sulfur carrier was added in the form of  $SO_4^{--}$ , and the target was dissolved in concentrated hydrochloric and nitric acids. Barium sulfate was precipitated from 1 N HCl. This was converted to barium carbonate by boiling with 50 percent aqueous potassium carbonate solution for about 1 hour. The precipitate was discarded, and the supernatant was made 1 N in hydrochloric acid. Barium sulfate was precipitated by the addition of barium chloride solution. The procedure was repeated twice. Finally, the barium sulfate precipitate was washed, dried at 110°C, and weighed as BaSO<sub>4</sub>.

#### 12. Phosphorus

Phosphorus was added as  $PO_4^{-3}$ , and the target was dissolved in hydrochloric and nitric acids. Phosphorus was then isolated by precipitation as ammonium phosphomolybdate from 1 N HNO<sub>3</sub>. The precipitate was dissolved in ammonium hydroxide, vanadium holdback agent was added, and magnesium ammonium phosphate was precipitated by the addition of magnesia mixture. The precipitate was dissolved in 1 N HNO<sub>3</sub> and the cycle was repeated. Finally, phosphorus was precipitated as magnesium ammonium 'phosphate. This was washed, ignited on a platinum dish, and weighed as  $Mg_2P_2O_7$ .

<sup>&</sup>lt;sup>3</sup> A. S. Newton, thesis, University of Michigan, Ann Arbor, Michigan (June, 1941), University Microfilm Service.

## 13. Silicon

Silicon carrier was added as  $(NH_4)_2SiF_6$ . After the target had dissolved, silica was brought down by the addition of boric acid and digestion with hot concentrated sulfuric acid. The silica was carefully washed with warm diluted sulfuric acid, then it was dissolved in potassium hydroxide. The solution was scavenged with titanium hydroxide. Holdback agents of iron, cobalt, chromium, vanadium, and yttrium were added, and the mixture was acidified with hydrochloric acid. Silica was precipitated by digestion. The cycle was repeated. The precipitate was then washed, ignited on a platinum dish, and weighed as SiO<sub>2</sub>.

# 14. Sodium

In the procedure sodium was left in ammoniacal solution. This solution was scavenged twice with iron and titanium hydroxides, with cobalt sulfide, and with calcium oxalate. The supernatant was acidified with hydrochloric acid and evaporated to dryness in a porcelain casserole. Ammonium salts were removed by fuming over a Meker burner. The residue was taken up in water, filtered, and zinc uranyl acetate was added to the cooled supernatant. The sodium zinc uranyl acetate precipitate was dissolved in alcohol saturated with hydrogen chloride gas and cooled. Sodium chloride precipitated. The precipitate was dissolved in a small amount of water, potassium holdback agent was added, and sodium zinc uranyl acetate was reprecipitated. The cycle was repeated once more. Finally, sodium chloride was washed with alcohol-hydrochloric acid, dried at 110°C, and weighed.

### C. Counting Techniques

The purified counting samples were transferred to weighed aluminum or platinum dishes and spread as uniformly as possible over the total area of the dish  $(\sim 5 \text{ cm}^2)$ . After suitable drying, the dishes were weighed again and the sample weight obtained by difference. The samples were then fixed in position on the dishes by evaporating dilute solutions of collodion or zapon on them, and the dishes were fastened to cardboard holders with Scotch tape, as shown in the diagram (Fig. 2). These holders were designed to fit in any one of five standard positions beneath the window of a chlorine-quenched Amperex Geiger-Müller tube. The position selected for counting a given sample was as far from the counter window as convenient, the criterion being the desirability of securing a sufficiently large number of counts in a short counting time to give good statistical reliability to the calculated counting rate. Absorption curves were taken with the absorber as close as possible to the counter window and the sample as far away as possible in order to minimize scattering of radiation into the counter by the absorber.<sup>4</sup>

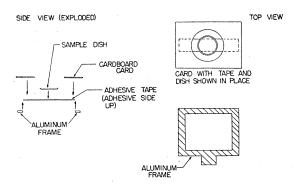


FIG. 2. Method used for mounting samples for counting.

The observed counting rates were subjected to the following corrections:

1. A correction for counts lost due to dead time of the counter tube (so-called coincidence correction).

2. A correction for background activity.

3. A geometric correction to the position in which the monitor sample was counted. This was determined empirically in each case.

4. A correction for air and window absorption, determined by back-extrapolation of the observed aluminum absorption curve.

5. A correction for backscattering from the sample dish. The figures used for beta-energies greater than 0.6 Mev were 1/1.22 for aluminum dishes and 1/1.75 for platinum dishes, calculated from the saturation backscattering data of Burtt.<sup>6</sup> For beta-energies less than 0.6 Mev the backscattering was calculated from the data of Yaffe and Justus.<sup>6</sup>

6. A factor for self-scattering and absorption by the sample material itself, estimated from the work of Malatesta.<sup>7</sup>

7. A factor for relative counting efficiency in the case of x-ray or gamma-activities. "Counting efficiency" is defined as the number of counts registered by the counter per particle (or photon) passing through the tube window.

8. A factor to correct for the decay of the sample from the time of the end of the bombardment to the time of initial measurement of the activity.

#### III. RESULTS

### A. New Chromium Isotope

In the chromium decay curves a new activity was found after subtraction of the activity due to 26.5-day Cr<sup>51</sup>. This half-life was found to be 19 hours, 24 hours, and 23 hours in bombardments FeP2, FeP3, and FeP5. Owing to a very poor chemical yield in bombardment FeP2, however, the last two figures should be more accurate. Since the electrons from Cr<sup>51</sup> can easily be blocked by a thin aluminum absorber ( $\sim 30 \text{ mg/cm}^2$ ), the growth and the decay of an isotope of a half-life of 16 days could be observed. In bombardment FeP5 two sets of absorption measurements were made, one after about 15 hours and the other several days after the bombardment. These measurements showed the growth of an activity with beta-radiation of about 0.7 Mev maximum energy. Furthermore, part of the purified chromium fraction from bombardment FeP5

<sup>&</sup>lt;sup>4</sup> R. C. Gibbs, Chairman, Division of Mathematical and Physical Sciences, National Research Council, Nuclear Science Series, Preliminary Report No. 8, "Conference on Absolute  $\beta$ -Counting," Papers 1 and 3 (October, 1950).

<sup>&</sup>lt;sup>5</sup> B. P. Burtt, Nucleonics 5, 32 (1949).

<sup>&</sup>lt;sup>6</sup>L. Yaffe and K. M. Justus, J. Chem. Soc. (London) S341 (1949), Part V.

<sup>&</sup>lt;sup>7</sup> L. Malatesta, private communication.

Р	ported in li	10001000		Experimen	ital results
Mass No.	Ti	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb (1 mb = $10^{-27}$ cm <sup>2</sup>
55	18.2 hr	β+, γ	FeP1 FeP2 FeP3 FeP5	18.3 hr 18.1 hr 17.6 hr 17.6 hr	0.42 0.14 0.53 0.53
56	72 days	β <sup>+</sup> , γ, EC	FeP3 FeP5	78 days 72 days	0.21 0.26

TABLE III. Yield of cobalt isotopes.

<sup>a</sup> Any previously reported data on half-lives or radiation characteristics is taken from G. T. Seaborg and I. Perlman, Revs. Modern Phys. 20, 585 (1948).

was left to decay for two weeks in the presence of inactive vanadium carrier. The vanadium was then separated chemically. This vanadium sample showed an activity with a half-life of 16.2 days and an energy of 0.78 Mev (determined from aluminum absorption measurements). The chromium isotope can from these experiments be assigned the mass number 48. Furthermore, the counting efficiency of the isotope was determined from the counting rate of its daughter to be

TABLE IV. Yield of iron isotopes.

Repor	ted in lite	rature	Experimental results			
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb	
52	7.8 hr	$\beta^+$	FeP1	8.1 hr	0.55	
			FeP2	8.3 hr	0.56	
			FeP3	8.3 hr	0.66	
			FeP4	8.4 hr	• • •	
			FeP5	8.3 hr	• • •	
			FeP6	8.3 hr	0.71	
			FeP7	8.6 hr	0.67	

about 4 percent which indicates that it decays mainly by electron capture.

#### **B.** Spallation Yields

## 1. Cobalt

It was assumed in determining the yield of  $Co^{56}$  that all long-lived activity was to be attributed to  $Co^{56}$ (see Table III).  $Co^{58}$  can only be formed from Fe<sup>57</sup> and Fe<sup>58</sup>, and these isotopes are in low abundance in natural iron (together 2.5 percent). This yield of  $Co^{56}$  is not corrected for any branching decay by electron capture.

TABLE V. Yield of manganese isotopes.

	Reported in	n literature	Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
51	46 min	β+	FeP3 FeP4	46.8 min 42.7 min	4.09 3.95
52	5.8 days	β <sup>+</sup> (35%), EC, γ	FeP3 FeP4 FeP5	5.8 days 5.8 days 5.7 days	14.5 11.0 13.1
54	310 days	β+, ΕС, γ	FeP3 FeP4		12.1 11.8
56	2.59 hr	β-, γ	FeP4 FeP4	2.72 hr 2.43 hr	0.56 0.61

## 2. Iron

In bombardments FeP4 and FeP5,  $Fe^{52}$  was used as a monitor (see Table IV). The counting efficiency of the isotope was determined by observation of the growth of  $Mn^{52}$  and by chemical separation of  $Mn^{52}$ . The counting efficiency was thus determined to be 1.72 for the equilibrium mixture. For  $Fe^{52}$  only, it is 0.70 which indicates that the parent isotope decays to the extent of about 30 percent by orbital electron capture.

TABLE VI. Yield of chromium isotopes.

Reported in literature			Experimental resu		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb
48	(Previously	unreported)	FeP2 FeP3 FeP5	19 hr 24 hr 23 hr	0.73 0.91 0.75
49	41.9 min	β+, γ	FeP2 FeP3 FeP7	41 min 40 min 43 min	4.9 6.1 3.5
51	26.5 days	EC, γ, e <sup>-</sup>	FeP2 FeP3 FeP5	(21 days) 27 days 28 days	53 38 33

# 3. Manganese

In the determination of the yield of  $Mn^{54}$  it was assumed that all the activity in the manganese fractions after the decay of  $Mn^{52}$  consisted of  $Mn^{54}$  (see Table V). The samples were observed for too short a time for a determination of the half-life. The figures for the yield are therefore uncertain. The counting efficiency of  $Mn^{54}$ was assumed to be 10 percent (from Miller's data).<sup>8</sup>

### 4. Chromium

For  $Cr^{51}$  a counting efficiency of 3 percent has been assumed (see Table VI). This figure is very uncertain, and the yield may be in error by a factor of two.

Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb
47	33 min	β+	FeP3	35 min	2.42
48	16 days	$\beta^+(58\%)$ , EC, $\gamma$	FeP3 FeP6	16.0 days 16.1 days	9 <b>.6</b> 11.0
49	600 days	ΕС, γ	FeP3	•••	31

### 5. Vanadium

The yield for  $V^{49}$  is only approximate since it was assumed that all activity after subtraction of 16-day  $V^{48}$ consisted of  $V^{49}$  (see Table VII). The counting efficiency for  $V^{49}$  is assumed to be 1 percent. (Decays solely by orbital electron capture.)

#### 6. Titanium

No long-lived titanium isotope was found (see Table VIII).

<sup>8</sup> D. R. Miller, AEC Declassified Document No. 2193 (August 6, 1948).

# 7. Scandium

The yield of  $Sc^{44m}$  was determined by counting through 222 mg/cm<sup>2</sup> aluminum absorber to block the radiations from  $Sc^{47}$  and  $Sc^{48}$  (see Table IX). It was assumed that the decay by orbital electron capture of  $Sc^{44}$  is negligible. The yield of  $Sc^{46}$  is not corrected for possible decay by orbital electron capture.  $Sc^{43}$  and  $Sc^{44}$ could not be separated in either bombardment nor could  $Sc^{47}$  and  $Sc^{48}$  in bombardment *FeP*6. Owing to the difficulty in separating  $Sc^{47}$  and  $Sc^{48}$  in bombard-

TABLE VIII. Yield of titanium isotopes.

Rep	orted in lit	erature	Expe	rimental re	sults
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb
45	3.08 hr	$\beta^+, \gamma$	FeP4 FeP7	3.13 hr 3.07 hr	3.31 4.05

ment FeP4 the figures for the yields are uncertain. Sc<sup>49</sup> could not be detected.

#### 8. Calcium

In *FeP*4 the decay of the calcium sample was observed for three months (see Table X). The decay follows a 152-day line fairly well. The absorption measurements made about two and one-half months after the bombardment (*FeP*5) indicated a  $\beta^-$  energy of 0.3 Mev and no gamma-radiation.

# 9. Potassium

The reason for the large difference in the yield determinations in the two bombardments may be due to occlusion of perchloric acid by the precipitate in bom-

TABLE IX. Yield of scandium isotopes.

Re	Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield mb	
43 and 44	both 3.92 hr	β+, γ, (EC)	FeP4 FeP6	4.1 hr 4.0 hr	$\begin{array}{r} 4.43\\ 4.14\end{array}$	
44	2.44 days	e <sup>-</sup> , γ	FeP4 FeP6	2.61 days 2.48 days	$2.61 \\ 2.59$	
46	85 days	β-, γ, EC	FeP4 FeP6	84 days	$3.27 \\ 3.14$	
47	3.4 days	β-	FeP4	3.5 days	0.84	
48	44 hr	β-	FeP4	44 hr	0.45	

bardment FeP6 (see Table XI). The higher figures for yield are therefore believed to be the more reliable.

# 10. Chlorine

The Cl<sup>34</sup> and Cl<sup>38</sup> activities were resolved by the use of a recording magnetic beta-spectrometer (see Table XII). The half-lives are obtained by the analysis of the data from this instrument, and they are not very accurate.

TABLE X. Yield of calcium isotopes.

Reported in literature		Experimental results			
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
45	152 days	$\beta^{-}$	FeP4 FeP5	(152 days) (152 days)	$\begin{array}{c} 0.54 \\ 0.58 \end{array}$
47	5.8 days	$\beta^-, \gamma$	FeP4 FeP5	5.6 days 5.5 days	0.007 0.006

#### 11. Sulfur

The sulfur decay was observed for about two months and showed a half-life fairly close to the reported one (see Table XIII). Aluminum absorption measurements showed a  $\beta^-$  energy of 0.15 Mev.

TABLE XI. Yield of potassium isotopes.

Rep	Reported in literature			imental re	esults
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
42	12.4 hr	$\beta^-, \gamma$	FeP6 FeP7	12 hr 12 hr	0.26 1.15
43	22.4 hr	$\beta^-, \gamma$	FeP6 FeP7	22 hr 21 hr	$\begin{array}{c} 0.12\\ 0.72\end{array}$

TABLE XII. Yield of chlorine isotopes.

Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb
34	33 min	$eta^+,\gamma$	FeP6 FeP7	36 min 27 min	0.08 0.14
38	38 min	β <sup>-</sup> , γ	FeP6 FeP7	45 min 37 min	0.13 0.20
39	55 min	$\beta^{-}$	FeP6 FeP7	55 min 58 min	$\begin{array}{c} 0.05 \\ 0.04 \end{array}$

TADIE	VIII	Vield	of sulfr	r isotopes.
LABLE	XIII.	Y 1eid	of sum	ir isolodes.

Reported in literature			Expe	rimental res	ults
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Vield, mb
35	87 days	$\beta^-$	FeP5	80 days	0.23

#### 12. Phosphorus

TABLE XIV. Yield of phosphorus isotopes.

Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
32	14.3 days	$\beta^-$	FeP5	15.0 days	0.044

## 13. Silicon

The half-life reported in literature seems to be too long. Both silicon samples were followed for a period of eight to ten half-lives, and both showed single component decay with a half-life of 157 minutes (see Table XV).

#### TABLE XVII. Summary of the yields observed.

#### 14. Sodium

The yield of Na<sup>22</sup> is uncertain because the decay was not followed long enough to establish the identity of the isotope with certainty (see Table XVI).

### IV. DISCUSSION

A cursory examination of the data presented in Table XVII seems to show no obvious systematic trend in the yields except that, as would be expected from

TABLE XV. Yield of silicon isotopes.

Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
31	170 min	$\beta^{-}$	FeP6 FeP7	157 min 157 min	0.12 0.125

previous work,<sup>9-13</sup> the nuclides formed in high yield are those which have atomic numbers only slightly below that of iron. The converse is not necessarily true, e.g., compare Cr<sup>48</sup>, 0.80 mb, with Sc<sup>44m</sup>, 2.60 mb. It is, however, possible to correlate the various yields somewhat more closely. To a first approximation natural iron can be considered to consist entirely of the isotope Fe<sup>56</sup> (91.6 percent).<sup>14</sup> At the bombarding energies under consideration the formation of a compound nucleus is believed to be unlikely. Transmission of excitation energy from the bombarding particle to the target nucleus is believed to take place by single nucleonnucleon collisions within the nucleus,15 the more energetic of the nucleons involved escaping from the nucleus. This will leave either Fe<sup>56</sup> or Co<sup>56</sup> in a highly excited

TABLE XVI. Yield of sodium isotopes.

Reported in literature			Experimental results		
Mass No.	$T_{\frac{1}{2}}$	Radiation	Bomb. No.	$T_{\frac{1}{2}}$	Yield, mb
24	14.9 hr	$\beta^{-}, \gamma$	FeP5	15.1 hr	0.026
22	2.6 hr	$\beta^+, \gamma$	FeP5	•••	0.021

state. Let us assume that these two species are formed in approximately equal abundance. Let us further assume either that protons and neutrons "boil off" with equal probability or that the most likely mode of dissipation of excitation energy is by emission of alphaparticles or deuterons with neutrons and protons boiling off with equal low probability. It can be seen that the most likely nuclide of a given mass to be formed by

Isotope	Vield (mb)	Remarks
Co <sup>55</sup>	0.49	
$\rm Co^{56}$	0.24	No correction for orbital electron capture
Fe <sup>52</sup>	0.68	· · · · · · · · · · · · · · · · · · ·
${ m Mn^{51}}$	4.0	
$Mn^{52}$	12.9	
$Mn^{54}$	12	10 percent counting efficiency. <sup>a</sup>
$Mn^{56}$	0.59	· · ·
$Cr^{48}$	0.80	
Cr <sup>49</sup>	4.8	
$Cr^{51}$	41	3 percent counting efficiency. <sup>b</sup>
$V^{47}$	2.4	
$V^{48}$	10.3	
$V^{49}$	31	1 percent counting efficiency assumed.
$Ti^{45}$	3.7	
$Sc^{44m}$	2.60	
$Sc^{46}$	3.20	No correction for orbital electron capture.
$Sc^{47}$	0.84	Uncertain.
$Sc^{48}$	0.45	Uncertain.
Ca <sup>45</sup>	0.56	
Ca47	0.007	
$K^{42}$	0.7	
$K^{43}$	0.4	
$Cl^{34}$	0.11	
$Cl^{38}$	0.17	
Cl <sup>39</sup>	0.045	
$S^{35}$	0.23	
$\mathbf{P^{32}}$	0.044	
$Si^{31}$	0.12	
Na <sup>24</sup>	0.026	
Na <sup>22</sup>	0.02	Approximate.

<sup>a</sup> See reference 8. <sup>b</sup> R. E. Batzel (private communication).

such a process must lie on a line represented on a plot of Z versus (A-Z) with a 45° (positive) slope passing through Co<sup>57</sup>. Such nuclides are those such that  $\tilde{Z} = 27 - \tilde{X}$ , A = 57 - 2X, where X = 0, 1, 2, etc. This simple picture would, of course, have to be modified for heavier nuclides, where the neutrons outnumber the protons appreciably. Some effects due to nuclear shell structure might also be observed.

Figure 3 represents a portion of the "Chart of the Nuclides" in which the ordinate represents the atomic number (Z) and the abscissa, the number of neutrons (A-Z) of the nuclides shown. The nuclides predicted by this simple picture to be formed in high yield are then those lying along the diagonal line marked "I" in

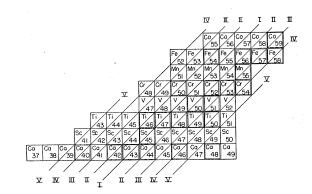


FIG. 3. Systematic grouping of nuclides formed by spallation of Fe<sup>56</sup>.

<sup>&</sup>lt;sup>9</sup> R. E. Batzel, Phys. Rev. 79, 528 (1950)

 <sup>&</sup>lt;sup>9</sup> R. E. Batzel, Phys. Rev. 79, 528 (1950).
 <sup>10</sup> H. H. Hopkins, Phys. Rev. 77, 717 (1950).
 <sup>11</sup> M. Lindner and I. Perlman, Phys. Rev. 78, 499 (1950).
 <sup>12</sup> N. A. Bonner and W. C. Orr, Phys. Rev. 76, 140 (1949).
 <sup>13</sup> Batzel, Miller, and Seaborg, Phys. Rev. 84, 671 (1951).
 <sup>14</sup> J. R. White and A. E. Cameron, Phys. Rev. 74, 991 (1948).
 <sup>15</sup> R. Serber, Phys. Rev. 72, 1114 (1947).

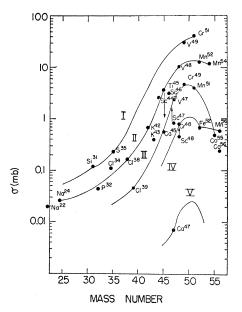


FIG. 4. Cross section for formation of various spallation products of natural iron as a function of mass number.

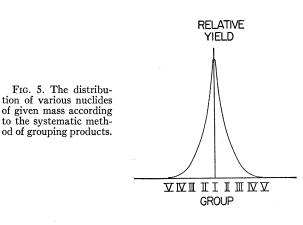
the figure; those next most probable by this model lie on the two lines marked "II," and so on. A tabulation of the expected nuclides is given in Table XVIII.

If one plots formation cross section *versus* mass number, it is apparent that all the nuclides of a given group (see Table XVII) fall on the same smooth curve with almost no exceptions and furthermore, that the curves so obtained are quite closely parallel on a semilogarithmic plot, as shown in Fig. 4. Since the curves are parallel on a semilogarithmic plot, the ratio of expected yields for two isobars on different curves is approximately constant. This fact enables us to draw from Fig. 4 an approximate distribution curve (Fig. 5) for the yields of various isobars of any given mass.

A few isotopes do not fall on their proper curves, e.g., for curve II,  $Sc^{46}$  is a little low. However, the counting efficiency of this isotope was not corrected for electron capture. Such a correction should indicate a higher value for the yield.  $Sc^{47}$  belonging to curve III is also low. This yield is very uncertain, however.

The yield of Ti<sup>45</sup> belonging to curve III is very high. This can be partially accounted for by considering that Fe<sup>54</sup> is in a rather high abundance (5.9 percent) in natural iron. With help of the distribution curve from Fig. 5 the contribution to the yields from Fe<sup>54</sup> has been estimated. It comes out to be 45 percent of the total yield for the isotopes  $Mn^{51}$ ,  $Cr^{49}$ ,  $V^{47}$ ,  $Ti^{45}$  but only 6 percent for  $Mn^{52}$ ,  $V^{48}$ , Sc<sup>44m</sup>. If the yields are corrected for this and also for the estimated contribution from Fe<sup>57</sup> and Fe<sup>58</sup>, Ti<sup>45</sup> falls below curve II even if it still seems to be high.

The picture given above indicates why Fe<sup>52</sup> is in



such low yield. It also explains why  $Sc^{49}$  (57 minute) could not be detected. It belongs to "curve" V and its yield is probably a factor of 100 lower than the yield of  $Sc^{44}$  (3.9 hour). It will therefore be completely masked by the latter isotope. Ti<sup>51</sup> also belongs to "curve" V. It could not be detected.

The yields of nuclides of mass 55 and above are not regular in this system. However, it is expected that these yields are low, since the nucleus is excited to a degree such that at least two or three nucleons may be expected to boil out. In this case the rarer, heavy isotopes of iron are expected to exert a disproportionately large effect of a magnitude which is difficult to estimate.

 TABLE XVIII. A systematic grouping of measurable product nuclides.

Group	Probability	Nuclides
I	Highest	Co <sup>57</sup> , Cr <sup>51</sup> , V <sup>49</sup> , S <sup>35</sup> , Si <sup>31</sup>
II	Next highest	Co <sup>56</sup> , Mn <sup>54</sup> , Mn <sup>52</sup> , V <sup>48</sup> , Sc <sup>46</sup> , Sc <sup>44</sup> , K <sup>42</sup> , Cl <sup>38</sup> , P <sup>32</sup> , Na <sup>24</sup>
III	Third highest	Co <sup>55</sup> , Mn <sup>51</sup> , Cr <sup>49</sup> , V <sup>47</sup> , Ti <sup>45</sup> , Sc <sup>47</sup> , Ca <sup>45</sup> K <sup>43</sup> , Cl <sup>39</sup>
IV V	Fourth highest Fifth highest	Mn <sup>56</sup> , Fe <sup>52</sup> , Cr <sup>48</sup> , Cl <sup>34</sup> , Na <sup>22</sup> Ca <sup>47</sup>

From Fig. 4 the total reaction cross section of natural iron for high energy protons is estimated to be 340 mb.

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