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Spallation Products of Arsenic with 190 Mev Deuterons

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March 21, 1949

Berkeley, California

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ABSTRACT

Purified arsenic has been bombarded with 190 Mev deuterons. Following chemical separation of chlorine and the 11 elements chromium through selenium, an analysis of the radioactivities in each fraction showed the presence of a total of 40 isotopes. An estimation of the yields of these isotopes has been made, and using the observed trends, yield figures were assigned to those isotopes which could not be observed. For a given value of Z (or A) the yields rise steadily to a maximum and then decrease; the line of maximum yields lies above the line of stability at masses 73 to 67, approaches it below. The yields of elements from arsenic to manganese decrease smoothly. Isotopes within a few mass units of arsenic must be formed by non-capture excitation processes, those farther away by capture processes followed by the emission of neutrons, protons and alpha particles. The chlorine isotopes are formed by the exclusive emission of alpha particles and neutrons.

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Spallation Products of Arsenic with 190 Mev Deuterons

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I. Introduction.

Since early 1947 the Chemistry Division of the Radiation Laboratory has investigated a type of nuclear reaction known as the "spallation" reaction[1]. This name applies to transformations by high energy particles (~100 Mev nucleons) which cause excitation of the target atoms to an extent that neutrons, protons, and alpha particles can be ejected. The product nuclei range from adjacent isotopes to isotopes lying twenty and more mass units lighter.

This paper summarizes an investigation of the spallation products formed when arsenic, $_{33}As^{75}$, undergoes bombardment by 190 Mev deuterons produced in the 184-inch cyclotron. The investigation involved the identification of the isotopes produced in the spallation, and the measurement of the yields of these isotopes. From the results the spallation can be described in terms of the numbers of charges and mass units emitted. The yield trends may be used as a basis for speculation concerning mechanisms of spallation.

II. Experimental Procedure.

A. Purification of Arsenic.

Preliminary experiments on the spallation of arsenic with 190 Mev deuterons[2] indicated that the products included nuclei of much lighter mass than the target nucleus; for example, Mn^{56} was identified. Since such isotopes are formed in a small yield (ca. 10^{-4} barns), it is necessary to know whether they could have been formed

from an impurity, rather than originating from a nuclear reaction on arsenic. The limits of impurities in the arsenic must therefore be known.

The arsenic at our disposal was "Kahlbaum" brand, reported spectrographically pure (greater than 99%). This material was sublimed in a pyrex tube. The middle fraction was resublimed in an electric furnace, and the sublimate collected on a thin aluminum foil, from which it was readily broken off. Spectrographic analysis of this twice sublimed material showed less than 0.1% impurities. To obtain greater sensitivity for the detection of impurities, forty milligrams of this arsenic were sublimed, and the residue was dissolved in hydrochloric acid for spectrographic analysis. The elements detected were Al 0.0005%, Ca 0.0001%, and Cu 0.0002%. The limits of detection for elements not observed are reported in Table I. The copper impurity found in the arsenic is too small to account for the formation of any of the observed isotope in the amount found. (The yields of isotopes from the spallation of copper are already known[3]). The distribution of chlorine isotopes observed indicates that they were not formed from potassium or calcium. No analysis was made for the non-metals sulfur, chlorine, argon and selenium. Chlorine and argon would have been removed in the sublimations. The absence of sulfur is indicated by the non-formation of Cl^{34}. There was no indication of elements above selenium.

Thus the arsenic was considered pure and the reactions we have observed are due only to nuclear transformations on arsenic.

Table I

Spectrographic Limits of Detection of Impurities.
Micrograms per 40 Milligrams Sublimed Arsenic.

K	1	Cr	1	Ni	0.1
Sc	0.01	Mn	0.1	Zn	0.1
Ti	0.1	Fe	0.1	Ga	0.1
V	0.1	Co	0.1		

B. Bombardment Procedure.

For the purpose of bombardment, several hundred milligrams of granular arsenic metal were wrapped with one mil platinum foil. The resulting envelope was approximately 25 mm long, 3 mm wide, and 2 mm thick. This envelope was clamped along one edge to a standard copper mount which is screwed to a water-cooled target head. This head moves into the cyclotron chamber so that the forward edge of the target meets the deuteron beam.

The duration of a bombardment was usually one hour. The beam attainable was about one microampere, but costly experience proved that the dissipation of the effective power caused volatilization of the target. Cutting the beam intensity to 30% eliminated this loss of arsenic as proved by weighings made before and after bombardment.

The energy loss of 190 Mev deuterons from penetrating one mil of platinum foil and 2 mm arsenic is 9 Mev. The reactions we are considering in this paper are not critical to this variation of energy, so we shall continue to speak as though we were bombarding a thin foil of arsenic with pure 190 Mev deuterons.

After bombardment, the envelope containing the arsenic was taken from the holder, and the arsenic removed. The pieces were ground in a hand mortar, and the resulting arsenic powder used for chemical operations.

C. Chemical Separations.

The separation of the isotopes produced by spallation into elemental fractions was accomplished by chemical procedures. First the target was dissolved in nitric acid. Second inactive carrier ions were added, corresponding to those elements we are concerned with. Finally, separations were conducted by distillations, solvent extractions, and precipitations.

The degree of separation required of the chemical procedures is determined by the radiation characteristics of the isotopes of the different elements and the relative intensities of the radiations. Thus, to identify conclusively two isotopes of different elements, of similar half-life, radiation properties, and yield, a separation of about fifty to one is required. In the case that the isotope of one element decays by electron-capture and the other by beta emission, then for the latter a separation factor of only five is required, due to the relative differences in counting efficiencies. Correspondingly, the elemental fraction of the electron-capturing isotope must be separated by a greater factor, of the order of 500, since small amounts of the beta emitter would introduce considerable error in counting the x-rays of the electron-capturing isotope. For cases where the half-lives differ considerably, other considerations must be applied. For example in the iron fraction only 47 day Fe^{59} is observed, while the longest gallium period is 3.3 days. Thus whereas gallium isotopes

are formed in far greater yield than Fe^{59}, no chemical separation is necessary, since all gallium isotopes will be dead before the Fe^{59} has decayed appreciably.

The quantity of a radioelement present is of the order of 10^{-13} mole. Thus, while this amount would undergo distillation and extraction just as milligram amounts of the element, most precipitations could not occur since solubility products would not be exceeded. Therefore, milligram amounts of the elements are added to function as carriers. The exchange of the radioactive isotopes with ions of the added carriers is believed to be complete in hot acid solution. No evidence such as cross contamination or widely varying specific activity indicated that this was not the case. Another reason for carriers is to minimize adsorption on precipitates.

A description of the separation of the elements into three groups will be presented, followed by a discussion of specific methods for individual elements.

1. The Germanium, Arsenic, Selenium Group.

These three elements may be distilled from 9N hydrobromic acid and bromine as the bromides. An alternative is to sublime the arsenic before dissolution in nitric acid. When this is done arsenic and radioselenium separate from the other radioelements. Germanium may subsequently be separated from the residue by distillation with hydrochloric acid.

This group may also be precipitated by hydrogen sulfide in acid solution and subsequently dissolved in ammonium hydroxide. The clear filtrate will contain only the thio-salts of germanium, arsenic, and selenium.

2. Iron and Gallium.

These elements are readily extracted into ether, from 5.5 to 6N hydrochloric acid. Iron must be in the ferric state. Washing the organic layer with more hydrochloric acid removes other groups which extract to a lesser extent.

3. Chromium, Manganese, Cobalt, Nickel, Copper, and Zinc.

This group can be separated from the first by the use of ammonium sulfide. The sulfides are usually dissolved in concentrated nitric acid in the presence of solid potassium chlorate, and subsequent procedures separate the elements one at a time.

Specific Procedures

1. Selenium.

The precipitation of selenium by reduction to the element with hydroxlamine hydrochloride is complete but slow in hot hydrochloric acid solution(1 to 4N). The reaction is specific among elements in this region, two precipitations sufficing to give a radioactively pure product. The reaction is accelerated by adding potassium iodide to 1N after the precipitation has started. Then the separation is quantitative in one minute, the product appearing as a liquid mixture of selenium and iodine. This can be dissolved rapidly in fuming nitric acid.

The selenium yield is determined by weighing the precipitated element.

2. Arsenic.

This element is generally separated as the sulfide from acid solution after removing selenium, and after removing germanium by distillation of the chloride. The arsenic sulfide precipitates slowly

but completely from ~3N hydrochloric acid. The reaction is accelerated by heating and vigorous agitation. If copper has not been removed, it will be separated when the arsenic sulfide is dissolved in ammonium hydroxide. Arsenic sulfide may be washed, dried and weighed.

3. Germanium.

Germanium tetrachloride distils readily from 6N hydrochloric acid. After boiling for six minutes, more than 95% of the germanium will be found in the distillate. The procedure is improved by utilizing a stream of chlorine. To completely separate from arsenic, the arsenic must be oxidized, which is easily accomplished by adding a few drops of chlorate solution. No arsenic will be found in the distillate. To recover the germanium, the chlorine in the filtrate is first destroyed with hydroxlamine. Passing hydrogen sulfide gas into the cold strongly acid solution precipitates white, flocculent, germanium sulfide. This precipitate may be washed, dried, and weighed.

4. Gallium.

The extraction into ethyl ether from 6N hydrochloric acid is complete ($>$ 98%) in two extractions. After four or five scrubs with one-third volumes of 6N hydrochloric acid the ether layer contains only iron and gallium. Separation of these elements can be accomplished by either of two methods, first the precipitation of ferric hydroxide away from the gallium in 1N sodium or potassium hydroxide, or second the reduction of ferric to ferrous ion followed by extraction of gallium from 6N hydrochloric acid. Ferrous iron does not extract. Gallium hydroxide may be precipitated at a pH of 5 in the absence of high concentrations of ammonium or acetate ion. For quantitative

determinations the precipitation of gallium 8-hydroxyquinolate was used. This precipitate can be washed with water, dried at 105° C and weighed.

5. Zinc.

The separation of zinc is based on the insolubility of zinc sulfide, or on the precipitation of zinc with 0.03M ammonium mercuric thiocyanate. Following the precipitation of the acid insoluble sulfide group from an acidity greater than 1N, zinc may be precipitated as the sulfide from 0.01M hydrogen ion. After dissolution of the precipitate, scavenging may be performed by precipitation of small amounts of ferric hydroxide from 1N potassium hydroxide. The zinc may then be reprecipitated as the sulfide from the alkaline solution.

Zinc may be removed from the original nitric acid solution by addition of the ammonium mercuric thiocyanate reagent to a 1M nitric acid solution. The precipitate is dissolved in concentrated nitric acid, inactive carriers are added for holdback purposes and the zinc reagent again added. The precipitate is white, granular, and easily filtered. It is readily washed and weighed on a filter paper.

Another useful property of zinc ion is its extractability into amyl alcohol from concentrated acidic thiocyanate solutions.

6. Copper.

The separation of copper is based on the precipitation of cupric sulfide and cuprous thiocyanate. Copper sulfide will be found in the precipitate containing the acid insoluble sulfides of selenium, arsenic, and germanium. Since copper forms no soluble thio compounds, the addition of ammonium hydroxide results in the dissolution of the

selenium, arsenic, and germanium compounds, leaving insoluble cupric sulfide behind. This compound is readily dissolved in nitric acid. After destroying the nitrate and reducing the cupric to cuprous with sulfite, white crystalline cuprous thiocyanate precipitates upon addition of ammonium thiocyanate to a solution of about 0.1M hydrogen ion. This compound or cupric sulfide may be weighed.

7. Nickel.

The precipitation of nickel dimethylglyoxime is quite specific. After separation of the germanium and gallium groups the alkali insoluble sulfides are precipitated as a concentration step. The precipitate is dissolved in nitric acid, the solution made ammoniacal, and the nickel reagent added. Two further precipitations from solutions containing holdback carriers suffice to produce a pure nickel fraction. The first precipitation is performed from ammoniacal solution, the second from acetic acid, acetate solution. The precipitate is easily washed, filtered, and weighed.

8. Cobalt.

The separation of this element is based on the extraction of the thiocyanate complex into amyl alcohol and the precipitation of potassium cobaltinitrite. Cobalt follows the separation scheme into the alkaline sulfide precipitate. This precipitate is dissolved in nitric acid and the nitrate destroyed by boiling down with hydrochloric acid. After the addition of 30% solid potassium thiocyanate, an equal volume of amyl alcohol is added. (Copper interferes.) Three or four extractions remove most of the cobalt, which is recovered from the alcohol by shaking with dilute base. After centrifugation the hydroxide is dissolved in acetic acid and potassium chloride

added. Potassium nitrite is then added until the precipitation of potassium cobaltinitrite is complete. This crystalline compound is easily filtered, washed, and weighed.

9. Iron.

The separation of iron depends on the extractability of ferric ion from 6N hydrochloric acid into ether. The ether layer is washed several times with 6N hydrochloric acid to remove traces of such elements as copper, arsenic, and germanium, which extract slightly. After extracting back into water, the iron is precipitated as ferric hydroxide from 1M hydroxide solution, the gallium remaining in the supernatant. This precipitation must be repeated at least four times, adding inactive gallium each time to remove all traces of gallium. The amount of gallium activity is initially five thousand times that of iron, so these steps are necessary.

Another separation from gallium is based on the non-extractability of ferrous ion into ether. Either stannous chloride or metallic mercury are satisfactory reducing agents for reducing ferric to ferrous ion.

A quantitative estimation of iron employs the spectrophotometric comparison of the absorption bands of the thiocyanate complex with those of standard solutions.

10. Manganese.

The precipitation of manganese dioxide separates manganese from neighboring elements. The alkaline sulfide precipitate is treated with hot fuming nitric acid, followed by potassium chlorate. The sulfides dissolve; after vigorous heating and agitation manganese dioxide precipitates. The dioxide dissolves instantly in acidic

peroxide solution. A by-product precipitation of arsenic and germanium sulfides must be performed to remove traces of these elements, which follow manganese dioxide precipitations. A reprecipitation of the dioxide from solutions containing inactive carriers will yield a pure manganese fraction. The dioxide is easily weighed.

11. Chromium.

The separation of chromium employs the ether extraction of the peroxychromate complex, H_3CrO_8. After treatment of the alkaline sulfide precipitate with nitric acid and chlorate, the chromium is in the VI state as dichromate ion. This solution is diluted to 1N acidity, cooled to 5° C and cold ether added. A drop of peroxide solution produces the intense blue color of the peroxychromic acid, which extracts into the ether. This ether layer must be washed several times with cold dilute nitric acid, to remove impurities. Dilute base removes the chromium from the organic layer. The chromate may be precipitated with lead or barium, and weighed. For further purification this precipitation is carried out in acetic acid solution.

12. Chlorine.

The volatility of chlorine is the basis for its separation. The dissolution of the metallic arsenic with nitric acid is performed in a still. A small amount of chloride ion is introduced with the nitric acid to function as a carrier. As the arsenic dissolves the chlorine is liberated as a gas and passes through the condensor into a water trap.

After reduction with hydrazine, the chloride is precipitated with silver and dissolved in 6N ammonium hydroxide. As a scavenging

step, silver iodide is precipitated from the ammoniacal solution. Upon acidifying the supernatant and adding more silver ion, the chloride precipitates. This precipitate can be weighed.

The above chemical procedures indicate the steps involved in the separation of these elements. In any given bombardment, one is usually interested in the complete separation of only three or four of the elemental fractions. Thus short cuts are used which by-pass groups of elements. To illustrate, suppose it was desired to separate manganese and nickel in pure form. One might first precipitate the alkali insoluble sulfide group, dissolve it with nitric acid and chlorate to precipitate the manganese dioxide, then dilute to a low acidity and precipitate nickel dimethylglyoxime. Of course, further purification steps are necessary.

The chemical procedures vary depending on the desirability of separating one particular element first, for example, to investigate a short half-life. Thus in order to observe 10 minute Cu^{62}, one would quickly separate selenium, reduce, and precipitate cuprous thiocyanate from the resultant solution.

A schematic separation of the elements investigates is shown in Fig. I, illustrating the basic separation steps. It should be realized that such a complete scheme would probably not be followed in any one bombardment.

D. Identification of Isotopes.

The identification of the isotopes depends on the measurements of radioactivity using a Geiger-Müller counter. Such features as the half-life, the energy of beta particles or gamma rays, the sign of beta particles, and the decay by electron-capture are determinable

experimentally and are used to identify individual isotopes.

The Geiger counters used in this laboratory are of the end-window type, approximately 4 inches long and 1 inch in diameter. The end is covered with a mica window of ca. 3 mg/cm^2 thickness. The tubes are filled to a pressure of 9 cm argon and 1 cm ethanol. These tubes operate at about 1200 volts and have a plateau of about 200 volts. The output pulses are fed into a scaling circuit of 64. The counting efficiencies of such tubes are 100% for beta particles and electrons, about 3% for 10 Kev x-rays, and 0.5% for 1 Mev gamma rays.

The half-life of a radioactive species is determined by counting its radiation at different times and plotting the counting rate on semi-log paper. The observed counts must be corrected for background radiation and coincidence counts. Usually the observed counting rate curve represents a mixture of two or more radioactivities decaying at different rates; the counting rate of the longer-lived activity is subtracted from the total curve, to yield the component due to the shorter-lived isotope.

The maximum energies of beta particles are obtained by counting with aluminum absorbers interposed between the sample and counter window, and thus determining the maximum range. A system known as Feather analysis can be applied to determine the energy relative to the known energy of some standard, such as P^{32}. The energies of electromagnetic radiations are obtained by determining the half-thickness for absorption in lead or aluminum. X-ray energies of elements in this region are determined by measuring the half-thickness in aluminum with a 1 gm/cm^2 beryllium absorber placed between source and counter, to stop electrons and beta particles, while only cutting the x-ray

intensity in half.

A simple beta-ray spectrograph has been used to investigate the radiation characteristics of various isotopes. This apparatus consists of a semicircular chamber placed between the pole pieces of a powerful electromagnet. Charged particles are bent around the semicircular path into a Geiger counter, according to their sign and energy. The magnetic field can be varied continuously and the counting rate recorded synchronously, yielding a plot of counting rate versus field strength. Thus the maximum energy of charged particles and the sign of the radiation can be determined. Another chamber permits direct counting of electromagnetic radiations, charged particles being deflected before they can reach the counter window.

The decay of a neutron deficient isotope by electron-capture (usually K-capture) is indicated by a high ratio of x-rays to electrons and beta particles. To verify that this is a decay process and not merely a K excitation, one can show that the x-ray energy is that of the daughter element. A simpler procedure is to show that there are no soft conversion electrons which might have originated from K-excitations. The use of the windowless counter "Nucleometer" is recommended for measuring the absorption of soft particles.

The measurements of the sign of a radiation and the half-life suffice to characterize most of the isotopes already known and reported in the literature. If an activity is observed which does not correspond to any that is known, the determination of the sign of radiation or the extent of decay by electron capture will place it on one side of the stable isotopes. To decide the correct mass number, one can look for active daughters of known mass number, or else per-

form cross bombardments.

E. Determination of Yields.

It is of interest to know the yields of isotopes formed in the spallation of arsenic with 190 Mev deuterons. The term "atom yield" refers to the number of atoms of a particular isotope formed in a bombardment. A more useful value, the "relative yield", refers to the ratio of the number of atoms of an isotope to the number of atoms of some one easily determined isotope. The yields of all isotopes are determined relative to this one. These figures are constant from bombardment to bombardment within experimental error. To obtain cross sections, it is only necessary to measure the cross-section for the formation of the standard isotope and multiply by each relative yield.

To determine a relative yield, the fraction of radioactive atoms of a given element which follow through the chemical procedure must be known. This value is taken equal to the fraction of added carrier recovered in the final solution. We assume complete exchange and no loss of activity before the addition of carrier.

The chemical recovery is determined by precipitating and weighing some compound of the carrier and comparing with the weight obtained from a solution of known concentration. The particular procedure employed has been indicated under the section on chemical separations.

After correcting for the chemical recovery, the number of radioactive atoms formed during bombardments must be calculated from the number of counts recorded by the Geiger tube at a later time. From the equation,

$$(1) \qquad N = \frac{-dN}{dt} \frac{T1/2}{0.693},$$

where N is the number of atoms at time t and $T_{1/2}$ is the half-life, $\frac{0.693}{\lambda}$,

it is seen that, by extrapolating the disintegration rate back to the midpoint of the bombardment, and multiplying by the half-life and constant 1.445, one obtains the number of radioactive atoms present at that time. If the decay has been appreciable(30%) during the time of bombardment, one must correct for those atoms which have decayed during that time. The correction factor is

$$(2) \quad F = \frac{t}{T\left[1-\exp\left(\frac{-0.693t}{T}\right)\right]},$$

where t represents the duration of bombardment, and T is the half-life.

Equation (1) involves the disintegration rate. To obtain the total disintegration rate from the sample counting rate, one must correct for: self-absorption, absorption, counting efficiency, and aliquot size.

Corrections for the absorption of beta particles in air and the counter window were made by obtaining absorption curves in aluminum and extrapolating to zero thickness absorber. This procedure assumes the absorption to be linear over the first six to eight mg/cm^2 of range. If the sample had a weight of several mg/cm^2, one-half this value was assumed to act as an absorber. No corrections were made for back-scattering, all samples being mounted on glass over cardboard or filter paper over cardboard.

Geometry corrections were applied using the relative counting rates of a UX_2 source placed at the different positions. Assuming 27% geometry for the first shelf, the factor for the second shelf is 10, for the third 21, for the fourth 38, and the fifth 55. Because of

variable scattering these factors should be determined for each radiation. For uniformity, our measurements were all obtained on the second shelf, where possible.

The counting efficiency of electrons and beta particles is assumed to be unity. For x-rays varying efficiencies were used according to the energies of the x-rays of the element concerned. For As the value 1.3% was used, for Ga 2.3%, Cu 3.5%, Co 5%, and Cr 8%. Dividing the counting rate of x-rays by the counting efficiency yields the number of x -rays per minute. In cases of orbital electron-capture it was assumed that each capture process is accompanied by one K x-ray.

In determining the yields of electron-capturing isotopes, it is necessary to divide by the fluorescent yield. This factor is the number of x-rays emitted per excitation. The values used are taken from Compton and Allison.[4]

III. Isotopes Formed in the Spallation of Arsenic with 190 Mev Deuterons.

The isotopes reported below have been found among the spallation products of arsenic with 190 Mev deuterons. An isotope is listed, followed by its reported mode of decay and half-life.[5] Next appears the observed half-life and yield relative to that of As^{72}, with an estimation of the probable error in the yield, either as a percentage or a factor. The estimated errors do not take into account inaccuracies due to incorrect assumptions of decay mechanisms or counting efficiencies. Those isotopes which were first observed in the course of this work are preceded by an asterisk.

Se^{75} K 127 day; Obs. 120 days, 0.14 (factor of 2).

This electron-capturing isotope was observed several months after bombardment, when the 9.5 day Se^{72} has decayed. The interfering 90 day As^{73} (from Se^{73}) must be separated chemically. The yield of Se^{75} was estimated by counting the x-rays in a more active sample from Se + D^{+}(190 Mev), and comparing the total Geiger activity in both samples.

Se^{73} β^{+} 6.7 hr; obs. 6.7 hr., 0.27($\pm$10%).

Measurements of the x-ray activity from this isotope indicate that the decay occurs by positron emission only 32% of the time. Decay of the positron was observed in the spectrograph; energy: 1.2 Mev.

Se^{72} K 9.5 days; obs. 9.7 days, 0.077($\pm$10%).

The positron from the 26 hour As^{72} daughter accounts for the activity decaying with the 9.5 day half-life. After chemical separation from arsenic, absorption measurements showed that the decay of Se^{72} is accompanied by less than 0.5% positrons or electrons.

Se^{69} β^{+} 44 min.; obs. 44 min. 0.004(factor of 2).

This isotope is formed in insufficient yield to observe directly; milking As^{69} from the selenium fraction indicates a half-life of 44 $\pm$ 4 minutes for Se^{69}. The yield is calculated from the amount of As^{69} grown in.

As^{74} β^{-}, β^{+} 17.5 days; obs. 19 days 1.25 ($\pm$10%).

We find a longer half-life of 19 days through a factor of 100. Both positron and negative beta particles are observed in the spectrograph.

As^{73} K 90 days; obs. long 2($\pm$ 50%).

This long period appears only after six months when the As^{74} is dead. Due to the low counting efficiency there are insufficient counts to determine an accurate half-life. We assumed a counting efficiency of 5%.

As^{72} β^+ 26 hr.; obs. 26.7 hr. 1.00(assumed standard).

The positron decay was observed in the beta-ray spectrograph. A search for x-rays indicated that decay by electron-capture occurs less than 2/3 of the time. Absolute cross-section measurements gave an average of 0.02 barns for the formation of As^{72}.

*As^{71} K; obs. 60 hr. 0.72($\pm$20%).

Between the 26 hour and the 19 day components of the arsenic fraction decay curve lies an unresolvable intermediate which we attribute to this isotope. The half-life and yield were obtained by milking 11 day Ge^{71} from the arsenic fraction. Seven milkings at 48 hour intervals yielded 60$\pm$4 hours for the half-life.

*As^{69} β^+ 52 min.; obs. 52 min. 0.14($\pm$ 50%).

Resolution of the 52 minute period is difficult due to the masking by As^{72}. The half-life is obtained from a sample of As^{69} milked from Se^{69}. The positron decay has been observed in the beta-ray spectrograph. Bombardments of Ga^{69} and Ga^{71} with helium ions of energy 10 to 32 Mev failed to produce this isotope; the mass must be less than 71, probably less than 70.

Ge^{71} K 11 days; obs. 11 days 2.0(±10%).

Decay of this isotope can only be seen by counting through 1 g/cm^2 beryllium, since otherwise it is masked by the positron of 68-minute Ga^{68} in equilibrium with 250 day Ge^{68}. The yield was computed from a count of the x-rays.

Ge^{69} β^+ 39.7 hr.; obs. 40 hr. 0.95(±10%).

From positron decay followed in the β-ray spectrograph we obtained 38.0 hours for the half-life. Measurement of the number of x-rays indicated that decay by positron emission occurs 27% of the time.

Ge^{68} K 250 day; obs. ~250 day 0.29(±20%).

This isotope is easily identified since the 68 minute Ga^{68} is soon in equilibrium. The gallium may be separated and the 68 minute decay observed, with a corresponding growth in the parent fraction.

Ge^{67} β^+ 23 min.; obs. 21 min. 0.12(±20%).

Ge^{66} ~140 min.; obs. ~150 min. 0.008(±20).

Resolution of the germanium fraction decay curve indicates a short period of about one hour. However, milking gallium daughters at regular intervals yields Ga^{67} and Ga^{66} in amounts corresponding to half-lives of 23 minutes for Ge^{67} and ~150 minutes for Ge^{66}. The one-hour period may be a combination of these. The yields of Ge^{66} and Ge^{67} are calculated from the amounts of the daughter activities.

If the germanium fraction is counted with an absorber of 1 gm/cm^2 beryllium, a half-life of 17 to 20 minutes is obtained. If the germanium fraction from Ge + D^+(190 Mev) is placed in the beta-ray spectrograph and the positron

decay followed, a 24 $\pm$ 3 minute half-life is obtained. Other activities are indicated.

Ga^{70} β^- 20.3 min.; obs. 20 min. 0.018(factor of 2).

This isotope is only observed by following the negative beta decay in the beta-ray spectrograph, since its resolution in a Geiger decay curve is masked by Ga^{68}. The yield is based on a comparison in the spectrograph of the total number of positive and negative particles counted in a sweep of the magnetic field.

Ga^{68} β^+ 68 min.; obs. 70 min. 0.56($\pm$20%).

The positron decay was followed in a Geiger counter and in the spectrograph. Measurements of the K x-rays indicated that less than 16% of the decay occurs by electron-capture.

Ga^{67} K 78.3 hr.; obs. 80 hr. 1.0($\pm$20%).

This longest-lived gallium isotope can readily be followed through a factor of 100. Absorption measurements gave the number of x-rays present, from which the yield was obtained, assuming 1.4 K-excitations per disintegration.(6)

Ga^{66} β^+ K 9.4 hr.; obs. 9.3 hr. 0.56($\pm$ 15%).

The half-life resolved from the Geiger decay curve was 11 to 12 hours, indicating a mixture of 9.4 hour Ga^{66} and possibly 14 hour Ga^{72}. Decay of the positive particles in the spectrograph was with a 9.3-hour half-life; of the negative, 12 hours. Absorption measurements on Ga^{66} from Cu + He^{++}(10 Mev) showed considerable x-rays; the fractional decay by positron emission was calculated to be 29%.

Zn^{72} β^- 49 hr.; obs. ~50 hr. 0.00022(±30%).

This period cannot be resolved from the zinc Geiger decay curve. Identification and yield estimation were accomplished by milking 14 hour Ga^{72} from the zinc fraction.

Zn^{69m} I.T. 13.8 hr.; obs. 13.5 hr. 0.015(±30%).

The decay is followed by counting the beta particles of 57 minute Zn^{69} in equilibrium.

Zn^{65} K 250 days; obs. long 0.16 (factor of 2).

A count of the x-rays indicated the yield of this isotope. The activity follows zinc chemistry and there are no other long-lived zinc isotopes.

Zn^{63} β^+ 38 min.; obs. 39 min. 0.022(±30%).

The Geiger decay curve shows a longer half-life, indicating a mixture with 57 minute Zn^{69}. Decay of the positron in the beta spectrograph gives a 38 minute half-life.

Zn^{62} K 9.5 hr.; obs. 10 hr. 0.0020(±30%) .

This isotope does not appear in the Geiger decay curve. The half-life may be observed by counting x-rays through 1 gm/cm^2 beryllium. The yield was calculated from the amount of 10.5 minute Cu^{62} separated at a given time.

Cu^{67} β^- 56 hr.; obs. 61 hr. 0.013(±10%).

Being the longest-lived activity in the copper fraction, this isotope is easily followed. Absorption and spectrograph measurement indicate a maximum energy of 0.55 Mev.

Cu^{64} β^-, β^+, K 12.8 hr.; obs. 12.8 hr. 0.20(±10%).

Both positive and negative particles are observed to decay with this half-life. We employed a recently determined ratio

of K excitations to positron disintegrations of 3.5 ± 1.0.[7]

Cu^{62} β^+, 10.5 min.; obs. 10 min. 0.10($\pm$10%).

A short bombardment time(ten minutes) enabled this isotope to be seen. Its decay was followed in the spectrograph.

Cu^{61} β^+,K 3.4 hr.; obs. 3.3 hr. 0.025 ($\pm$10%).

The decay of this isotope could be followed in the spectrograph by adjusting field strength so that only the most energetic particles were counted. The extent of electron-capture is not known.

Ni^{66} β^-, 56 hr.; obs. 56 hr. 0.006($\pm$20%).

This isotope is identified by milking the 10.5 minute Cu^{66}. It is the longest period in the nickel fraction.

Ni^{65} β^-, 2.6 hr.; obs. 2.63 hr. 0.0030($\pm$20%).

The 2.6 hour period is seen as soon as the nickel fraction is separated and may be followed for several half-lives. The negative beta particles were observed in the spectrograph.

Ni^{57} β^+ 36 hr.; obs. $\sim$34 hr., 0.00003($\pm$50%).

This isotope can only be observed by following the decay of the positrons in the spectrograph. Since the activity level is quite low, only an approximate value of the half-life is obtained. By comparing areas of the positron sweep curve and the negative beta sweep curve, the yield of Ni^{57} relative to Ni^{66} is obtained. The extent of electron-capture is not known.

Co^{61} β^-, 1.75 hr.; obs. 1.83 hr., 0.014(±40%).

The decay of this cobalt activity may be followed through a factor of ten. The negative beta particles have been observed in the spectrograph.

$Co^{56,58}$ β^+,K 72 days; obs. long, 0.006(factor of 2).

The long-lived cobalt activity is probably Co^{58}. The Geiger decay curve indicates a decay period of ~90 days which apparently turns into a longer-lived component, Co^{57}. Absorption curves yielded the number of K x-rays in the sample. Using the reported positron ratio, 15%, we obtained the above relative yield.

Co^{55} β^+, 18.2 hr.; obs. ~18 hr., 0.00022(±50%).

A Geiger decay curve of the cobalt fraction does not show this period; it can only be seen by counting positrons in the beta-ray spectrograph. The level of activity is too low to obtain an accurate half-life. The yield was obtained by comparing the areas under the sweep curve of the positrons from Co^{55} with the area under the sweep curve of the negative beta-particles from Co^{61}.

Fe^{59} β^- 46.3 days; obs. 46 days, 0.005(±30%).

This is the only period seen in the iron fraction. A rapid separation from gallium was performed in an attempt to observe 8.9 minute Fe^{53}. The yield of this isotope must be less than 0.00005. The yield of Fe^{52} is less than 0.000005.

Mn^{56} β^- 2.59 hr., obs. 2.6 hr. 0.0026(±30%).

This is the shortest period seen in the manganese fraction. There is no evidence for 46 minute Mn^{51}. The decay may be followed through a factor of ten.

Mn^{52} β^+,K; 6.5 days; obs. 6.2 days 0.0003(±30%) .

The level of activity from this isotope is quite small. Using the reported decay ratio of 35% positrons, an overall efficiency of 40% was assumed. It is not known how much of the Mn^{52m} is formed.

Cr^{51} K, 26.5 days; obs . 25 days 0.002(±50%).

The level of activity observed from a one hour bombardment is quite small, ≈50 c/m. An overall counting efficiency of 8% was assumed.

Cr^{49} β^+ 41.9 min.; obs. 40 min. 0.00006(±40%).

This is the only short-lived component in the chromium decay curve. No intermediate period is seen corresponding to a 1 to 2 hour Cr^{55}. The extent of electron-capture is unknown.

Cl^{39} β^- 1 hr.; obs. ≈ 53 min., 0.00001(±50%).

Cl^{38} β^- 38.5 min.; obs. ~ 38 min., 0.00002(±50%).

The decay of the chlorine fraction bends away from a 40 minute line, and keeps turning until a long tail is reached. The only way to resolve the 38 min. line is to assume a 53 min. component is also present. The yield of this component, 0.00001, coupled with data concerning the known chlorine isotopes, places this period at Cl^{39}. A similar effect is observed in the chlorine fraction removed from [Cu + D^+(190 Mev)] [13].

A summary is presented in Table II, listing the observed isotopes, their decay mode, the literature half-life, the observed half-life, the observed relative yield and the charge, mass difference from $_{33}As^{75}$.

Table II

Isotopes Produced by $_{33}As^{75} + D^{+}$(190 Mev).

Isotope	Decay Mode(5)	Literature Half-Life(5)	Observed Half-Life	Yield rel. As^{72}	Difference
$_{34}Se^{75}$	K	127 days	120 days	0.14	-1z,0a
Se^{73}	β^+,K	6.7 hr.	6.7 hr.	0.27	-1z,2a
Se^{72}	K	9.5 days	9.7 days	0.077	-1z,3a
Se^{69}	β^+	44 min.	~44 min.	0.004	-1z,6a
$_{33}As^{74}$	β^+,β^-	17.5 days	19 days	1.25	0z,1a
As^{73}	K	90 days	(long)	2	0z,2a
As^{72}	β^+	26 hr.	26.7 hr.	1.00	0z,3a
As^{71}	K	60 hr.	60 hr.	0.72	0z,4a
As^{69}	β^+	52 min.	52 min.	0.14	0z,6a
$_{32}Ge^{71}$	K	11 days	11 days	2.0	1z,4a
Ge^{69}	β^+,K	39.7 hr.	40 hr.	0.95	1z,6a
Ge^{68}	K	250 days	~250 days	0.29	1z,7a
Ge^{67}	β^+	23 min.	21 min.	0.12	1z,8a
Ge^{66}	β^+(?)	140 min.	~150 min.	0.008	1z,9a
$_{31}Ga^{70}$	β^-	20.3 min.	20 min.	0.018	2z,5a
Ga^{68}	β^+	68 min.	70 min.	0.56	2z,7a
Ga^{67}	K	78.3 hr.	80 hr.	1.0	2z,8a
Ga^{66}	β^+	9.4 hr.	9 hr.	0.56	2z,9a
$_{30}Zn^{72}$	β^-	49 hr.	~50 hr.	0.00022	3z,3a
Zn^{69m}	I.T.	13.8 hr.	13.5 hr.	0.015	3z,6a
Zn^{65}	K	250 days	(long)	0.16	3z,10a
Zn^{63}	K	38 min.	39 min.	0.022	3z,12a
Zn^{62}	β^+	9.5 hr.	10 hr.	0.0020	3z,13a
$_{29}Cu^{67}$	β^-	56 hr.	61 hr.	0.013	4z,8a
Cu^{64}	β^+,β^-,K	12.8 hr.	12.8 hr.	0.20	4z,11a
Cu^{62}	β^+,K	10.5 min.	10 min.	0.10	4z,13a
Cu^{61}	β^+	3.4 hr.	3.3 hr.	0.025	4z,14a

Table II(cont'd)

Isotope	Decay Mode(5)	Literature Half-Life(5)	Observed Half-Life	Yield rel. As^{72}	Difference
$_{28}Ni^{66}$	β^-	56 hr.	56 hr.	0.0006	5z,9a
Ni^{65}	β^-	2.6 hr.	2.6 hr.	0.0030	5z,10a
Ni^{57}	β^+	36 hr.	34 hr.	0.00003	5z,18a
$_{27}Co^{61}$	β^-	1.75 hr.	1.83 hr.	0.014	6z,14a
Co^{58}	β^+,K	72 days	long	0.006	6z,17a
Co^{55}	β^+	18.2 hr.	18 hr.	0.00022	6z,20a
$_{26}Fe^{59}$	β^-	46.3 days	46 days	0.005	7z,16a
$_{25}Mn^{56}$	β^-	2.59 hr.	2.6 hr.	0.0026	8z ,19a
Mn^{52}	β^+,K	5.8 days	6.2 days	0.0003	8z ,23a
$_{24}Cr^{51}$	K	26.5 days	25 days	0.002	9z,24a
Cr^{49}	β^+	41.9 min.	40 min.	0.00006	9z,26a
$_{17}Cl^{39}$	β^-	1 hr.	~53 min.	0.00001	16z,36a
Cl^{38}	β^-	38.5 min.	38 min.	0.00002	17z,37a

IV. Discussion of Results.

Introduction.

A high energy deuteron approaching an arsenic nucleus may be pictured as a dumbbell shaped object - moving at high speed(~0.4c) towards a spherical assemblage of nucleons. In a collision with the arsenic nucleus, the relatively weak deuteron binding will be broken. The effect is that of two high energy particles, a neutron and proton, approaching an arsenic nucleus together.

If the deuteron approaches the arsenic nucleus at the edge so that only one of its nucleons will collide with the arsenic, this

nucleon is effectively separated from the other, which continues on its line of flight. This process is termed stripping by Serber[8]. The energy of the stripped component may vary from 75 to 115 Mev.

Should the whole deuteron collide with the nucleus, there are collisions and resultant excitations due to both particles.

Serber's picture of high energy reactions incorporates the phenomenon of nuclear transparency[9]. High energy particles have a mean free path of the order of the nuclear diameter. Thus some incident particles may pass through the nucleus without collision. Other particles may undergo one or two collisions near the edge and then recoil out of the nucleus.

Thus we have the possibility of excitations over a wide range of energy, from 190 Mev when the whole deuteron strikes a nucleus, and all its energy is dissipated in collisions, to small excitations of 25 Mev when one nucleon suffers a surface collision and then recoils outward.

The yields of spallation products of arsenic bombarded with 190 Mev deuterons indicate only the relative occurrences of processes leading to a certain net change in Z and A. However, to reach a given nucleus requires a minimum mass energy and barrier energy. Thus the frequencies of processes requiring excitations greater than various chosen values can be estimated. If certain isotopes are found to have low yields, it is inferred that either the requisite excitation is unlikely, or the particular process leading to the isotope is unlikely.

A. Trends in the Observed Yields.

The yields of isotopes observed in the spallation of arsenic with 190 Mev deuterons have been plotted in Fig. II.

Yields of stable isotopes cannot be measured, and very short-lived or very long-lived isotopes could not be observed under the conditions of our bombardments. Thus out of some 90 isotopes contained in the stable region from chromium through selenium, we have measured yields of only 40, or about 44%, of which nearly half are found among the top four elements. This inadequate representation must be kept in mind.

1. For each element there is a smooth rise and fall of yield as the mass number is varied. This trend holds for each element of which the yields of several isotopes have been measured, that is for the seven elements nickel through selenium. There is no evidence for any marked periodicity with mass number.

2. For each isobaric series, the yields increase smoothly towards the region of stability. This region can be thought of as a band including the center of the stable triad of each even element, and the surrounded atom of each odd element dyad. The rule holds for eight cases below selenium, including masses 52 and 65, for which limits on the yields of Fe^{62} and Ga^{65} were obtained. These cases are all on the neutron deficient side; there are no examples on the heavy side.

3. Yields decrease as one progresses from germanium to lower elements. Looking at the highest yield observed for gallium, copper, cobalt, manganese, and chlorine, we find 1, 0.2, 0.01, 0.003, 0.00002. The trend holds for the even elements as well. Nickel is an apparent exception, but is discounted since no measurement was made over a region of seven isotopes, including the stable ones.

4. Among the elements above copper the yields of neutron deficient isotopes predominate, below copper the heavy isotopes are formed in greater abundance. The yield of 0.02 for Cr^{51} indicates an exception to this rule, as Cr^{55} is not observed. However, the assumed counting efficiency of Cr^{51} may be low(8%), and the reported characteristics of Cr^{55} are still open to doubt.

5. The yields of selenium isotopes are smaller than those of the corresponding arsenic isotopes by factors of the order of ten.

B. Trends in the Complete Yield Diagram.

It is desirable to interpolate (and extrapolate) yields of the isotopes which were not observed. This has been done, using the principle that yields for a given Z shall rise and fall smoothly. Examining the region copper through selenium, it appears that the yields of adjacent isotopes near the maximum value for a given element vary by a factor of 1.5 to 2, several mass units out by a factor of 3, and farther by 10. These ratios have been applied to the observed yield figures to obtain the yields of neighboring isotopes.

Fig. III includes the interpolated yield values as well as the measured ones. What trends are to be found?

1. For each Z there is a smooth rise and fall of yield with A. The chart was so constructed.

2. For each isobaric series the yields increase smoothly towards a line lying one mass unit above the line of stability from masses 66 through 71 and approaching the line of stability at lower masses. This line has been drawn in Fig. III. In all cases but two the yields increase as one approaches the line from either side. This line indicates the most probable region for the formation of spallation

products of a given mass number.

If the yields could be plotted perpendicular to the paper and the resulting yield surface smoothed, we would have a ridge running along the line of most probable formation and steadily diminishing in height. Near the peak, (germanium and arsenic), the ridge breaks off sharply to the right but decreases gradually to the left.

The maximum yield of an isotope of given Z lies adjacent to this line, or at the top of the ridge. This behavior can be clearly seen at gallium and below.

3. Yields decrease as one moves along the isotope region from germanium to lower elements. This effect is an overall trend, and is seen best by comparing yields along the line of most probable formation.

4. The yields of neutron deficient isotopes predominate above copper; below copper the high yields for a given Z shift towards the stable and neutron excess region. At chlorine we observe only Cl^{38} and Cl^{39}, isotopes with a neutron excess.

The total yield of isotopes with a given mass number is plotted against the mass number in Fig. IV. A smooth curve without inflection is drawn from mass 76 to 56 and all points lie within ~30% of their value from this line. The apparent rise at mass 55 is not understood. Considering the inaccuracies of yield measurements as well as those of the yield interpolations, there are no erratic yield deviations that indicate preferred isotope formation.

The total yield for a given atomic number Z is plotted against the atomic number in Fig. V. The deviations from a smooth curve are more pronounced. The hump at chromium (Z = 24) may not be real, since

90% of the yield is due to the extrapolated values, and because of the uncertainty in the counting efficiency of Cr^{51}.

C. Comparison of Selenium and Arsenic Yields.

The formation of selenium isotopes can only occur through the capture or exchange of a proton, and subsequent emission of neutrons. The arsenic isotopes can be formed by excitation without capture, or by capturing a proton and then emitting a proton and neutrons, or by capturing a neutron and subsequently emitting neutrons. If for example neutron and proton reactions on arsenic are equally likely, and the emission of one, two or three particles (neutron or proton) is equally probable, then arsenic isotopes should be formed twice as often as selenium. Actually neutrons are somewhat more likely to be emitted, as shown by the yield distribution maximum lying to the left of the stability line. Since neutrons are more likely to be emitted than protons, following equally probable proton or neutron capture collisions with arsenic nuclei, one would expect the ratio of arsenic to selenium yields to be less than two for the above example. Since this is not the case it is believed that the reactions leading to the arsenic isotopes in this region result from a different mechanism, namely non-capture excitations in which the high energy nucleons impart a portion of their energy to the nucleus and then proceed.

The selenium isotopes are formed from less likely processes which involve capture of a charge followed by deexcitation with the emission of only a few neutrons. Such reactions include the stripping of a proton by the nucleus and capture of the proton, an exchange reaction with this proton, or complete capture of the deuteron.

Since the frequency of these processes is thought to be at least of the same order of magnitude as that of non-capture excitations, these processes mus t involve high excitations (100 to 180 Mev) ordinarily leading to isotopes of elements lying lower in the table. That is to say, the resultant high excitation makes the emission of at least one charged particle a likely event.

To summarize, arsenic isotopes are formed from non-capture excitation processes. The low selenium yields are due to the improbability of capturing a charge and causing only a low excitation.

D. Energy Requirements.

A discussion of the yields of isotopes below arsenic and selenium is enhanced by a knowledge of the energy required to effect a given change in Z and A. Energy is needed to make up mass differences, and to overcome the potential barriers.

In the arsenic region binding energies are approximately 9, 8, and 3 Mev for neutrons, protons, and alpha-particles. In addition, 5 Mev coulomb energy must be added per charge. Thus to reach Ga^{70} by emitting neutrons and protons we estimate 53 Mev is required, for Cu^{66} 100 Mev, for Ni^{61} 150 Mev, and for Fe^{56} 190 Mev. These are rough figures and neglect kinetic energy of the outgoing products. Nevertheless it becomes apparent that below mass 56 (190 Mev) alpha emission must account for a large proportion of the reactions, since these reactions are energetically impossible otherwise. The estimated yield of all species below mass 55 is only 0.3% of the total yield so the many alpha-particle emission is a relatively rare event.

A more exact treatment utilizes exact mass differences, and estimates only the binding energy for isotopes adjacent to those whose

masses are known. Using an arsenic mass 74.947 calculated from the mass equation(10), the formation of Cl^{38} from As^{75} by emitting alphas and neutrons requires 103 Mev in mass energy and ~65 Mev to overcome the barriers. The coulomb barrier was estimated from the expression $\frac{2zZe^2}{3r}$.(11) Thus the formation of Cl^{38} requires 168 Mev out of a maximum possible excitation of 190 Mev.

The remaining 22 Mev can be distributed as kinetic energy among the 8 alpha particles, 5 neutrons, and the outgoing deuteron components. These particles therefore must be evaporated with a mean kinetic energy less than 2 Mev in excess of energy required from passing over barriers. This particular process of deexcitation with the emission of nearly the maximum possible number of alpha particles is a rare event, accounting for only 10^{-6} of the total yield.

From the exact mass differences and full barrier height it is seen that the minimum energy required to reach mass 60 by the emission of alpha particles and neutrons is ~70 Mev, to reach mass 55, ~100 Mev. The region of greatest abundance (93% of products) from Ga^{66} to As^{75} requires excitation of 10 to 65 Mev (kinetic energy excluded). If only (excitation,n,p) reactions are considered the minimum excitation for this region of greatest abundance ranges from 10 to 80 Mev. The next group of isotopes from A = 56 to 65 requires minimum excitation of 70 to 110 Mev, if as many alphas are emitted as possible. These minimum requirements are indicated in Fig. VI. Fe^{57} is the lowest isotope which can be reached through the evaporation of just neutrons and protons.

The difficulty with correlating excitation energies and yield data is that any given excitation will probably not be followed by a deexcitation leading to only one isotope.

E. Mechanism.

It has been shown that the arsenic isotopes are probably formed from non-capture excitations by the deuteron, followed by neutron emission. This mechanism should hold for the formation of germanium and gallium isotopes, the emitted particles including a proton or alpha particle.

Beginning at the gallium-germanium region and moving down the table, we expect contributions to the yield due to processes involving complete capture of the deuteron by the arsenic. Isotopes such as Cr^{51} (minimum excitation 132 Mev) are formed from such a process. With the nucleus at high states of excitation the probability that an emitted particle be a proton is always somewhat greater than the probability that it be an alpha. Thus below copper we expect a smooth decrease in yield from element to element, since in a stepwise deexcitation the probability that one of the emitted particles be a proton becomes quite large. The yields should gradually shade off to the one element below which it is energetically impossible to go. The yields immediately above this element will be concentrated on the neutron excess side.

The deexcitation process occurs in a stepwise fashion, roughly following the line of most probable isotope formation as the various particles are evaporated. The stepwise path is implied in the fact that the yields decrease away from this line. If for example all the neutrons came out first we should expect larger yields on the neutron

deficient side of the lighter elements than on the neutron excess side, which is not the case. Also shielded isotopes are formed, the best example being that of Cu^{64}, surrounded by stable isotopes.

F. Comparison with Theoretical Calculations.

Horning and Baumhoff have examined the problem of high energy deuteron bombardment of a nucleus A = 100 in an attempt to predict the prong distribution of stars formed in emulsions (12). They have calculated the probability that a 200 Mev deuteron cause an excitation X in the range dX in a collision with a nucleus of A = 100. This curve rises steadily to 100 Mev, drops down, and then rises to 200 Mev. The drop at 100 Mev is over a factor of 3, the subsequent rise over a factor of ~ 4. The most likely excitation is just below 200 Mev, the next most likely below 100 Mev. In the yield curves of the spallation products of arsenic we see no such pronounced rise and fall; a given excitation does not lead to only one mass number or element.

Since 85% of the yield is between Ga^{66} and As^{74}, Horning and Baumhoff's curve indicates that excitations of the order 140 to 190 Mev(and 40 to 95 Mev) have led to isotopes in this region. Since these isotopes are formed by the emission of one to 9 particles but require less than 70 Mev excitation, on the average the emitted particles must carry off considerable kinetic energy, some up to ~ 30 Mev apiece.

Thus the correlation of yields with minimum required excitations has little significance, since most paths of deexcitation use up more than the minimum energy for that path.

In Fig. IV the yields of masses 66 and 67 (due chiefly to Ga^{66} and Ge^{67}) are higher than a smooth fit to the points by about 40%.

This effect is open to some question due to the inaccuracies of the data and extrapolations. The isotopes at these masses, requiring 60 to 70 Mev excitation, appear to correspond to Horning and Baumhoff's central peak where a maximum excitation is found from 60 to 100 Mev due to the stripping reactions. From their curves showing the probability that a given evaporated particle be an ion[13] we see that the formation of an ion is only 1/10 as probable as a neutron. Since the probability that a given ion be an alpha is small[13] (excitation 100 Mev) most of the reactions due to these excitations would involve emission of neutrons and protons. The requisite energy to reach masses 66 and 67 is about 80 Mev, and assuming an average of 2 Mev kinetic energy per particle we should need a total of 96 Mev. Thus excitations from this portion of Horning and Baumhoff's curve <u>can</u> lead to Ga^{66} and Ge^{67}. However since 96 Mev is very close to the limit of their peak(100 Mev for a <u>200</u> Mev deuteron), and since there is a spread of kinetic energy of a nucleon emitted from a nucleus of given excitation, it is unlikely that this hump at masses 67 and 66 is a real effect.

Horning and Baumhoff have calculated the probability that an ion evaporated from a nucleus of excitation X be a proton[12]. From 100 to 190 Mev this probability is $\sim$0.5, at 30 Mev it is practically unity. For high excitation the probabilities are roughly the same that one of the first few ions evaporated by a proton or alpha; for lower excitations the emission of protons is favored. These results also lead us to expect a smooth variation of yield with Z, without pronounced oscillation, as mentioned before. When a nucleus has been deexcited to 30 Mev, there is little chance that further emission

of alpha particles or protons can occur, so the last particles emitted are most likely to be neutrons.

Horning and Baumhoff estimated the distribution of prongs from stars initiated in photographic emulsions by 200 Mev deuterons[12]. The calculated mean prong number is 4.4, of which only $\sim$ 3 will be observed in the emulsion. (This value is in agreement with the experimental work).[14] About 95% of the stars should have prong numbers equal to or less than 6. If we consider the arsenic spallation product yields, and interpret the yield for a given change in atomic number greater than one as indicating the number of theoretical stars of given prong number, then the prong number for 95% of the spallation "stars" is less than or equal to 4, with a mean of 2.8. For Z = 29 and 30 the probability is greater that the arsenic atoms captured the deuteron and the extra proton should count as a prong. The range of prong numbers is then less than 5 and the mean is 26, on the new scale. Here is a discrepancy of $\sim$1.6 or 1.8 between the predicted mean prong number and that inferred from the arsenic spallation yields. The discrepancy becomes greater when we consider that some changes of ΔZ are due to alpha emission.

In their article, Horning and Baumhoff present a table listing the number of stars of different prong number observed in the cloud chamber when a 90 Mev neutron beam is passed into water and alcohol vapor. The overall shape of this distribution is similar to that of the arsenic "stars", considering $\Delta Z = 2$ as a two pronged star. Only qualitative agreement is to be expected, considering the differences in target atoms.

G. Comparison with Copper and Antimony Spallation Yields.

The general yield pattern of the copper spallation products[3] is the same as for arsenic. One difference is that the yields from copper do not drop off as fast on the neutron deficient side, as those of arsenic do. Thus six elements removed from the target, the yields from arsenic are about 10 times smaller than those of the corresponding isotopes from copper. Another difference is found in manganese isotopes from copper, the yield of Mn^{54} is reported about 50% smaller than either Mn^{52} or Mn^{56}, whereas from our work we would expect it to be larger.

The "plateau" found in the copper spallation yields over the range of masses 50 to 59 does not show up in the corresponding arsenic region, 61 to 70. Here we have a general decline in yield, through a factor of 40. Although yields of elements with odd Z are somewhat higher in the case of copper spallation products, there is no good evidence for this in the case of arsenic.

The distribution of the antimony spallation product yields[15] is also similar to that of arsenic. In the case of the antimony region there is a much broader range of stability for a given Z, as shown by the greater number of stable isotopes for the even elements. Thus the yield curve of isotopes of given Z appears to be much flatter than in the case of arsenic spallation products. For Z = 44 we find yields of Ru^{97}, Ru^{105} and Ru^{106} all within a factor of 5. In the case or arsenic spallation products we find a variation of a factor of 1000 from Fe^{53} to Fe^{59}, the comparable case.

Two specific differences may be mentioned. Among the antimony spallation products are Ag^{105} and Ag^{106} which are formed in yields

more than 100 times as large as the corresponding extrapolated yields of Cu^{58} and Cu^{59} from arsenic spallation. In the antimony case however, these isotopes lie very close to stability, which is not true of the copper isotopes. On the neutron excess side, an exceptionally high yield is observed at Pd^{111}, and a yield 1/100 of this is found at Pd^{109}, which is situated between stable isotopes. There is no corresponding case among the arsenic yields.

H. Problems in Arsenic Spallation.

Two features of the arsenic spallation yield distribution are not understood. These are connected with the yields of the neutrom deficient isotopes of chromium, manganese, and iron. Although Co^{55}, Mn^{52}, (and Cr^{49} and Cr^{51}) are detected, no trace of Fe^{53} was found, the limit being less than 1/10 the yield of Cr^{49}, which is just one alpha particle removed.

The other anomaly is the apparent high yield of Cr^{51}, leading to the discontinuities in the curves of yield vs. mass number and vs. atomic number. To repeat, the uncertain counting efficiency of Cr^{51} may be the trouble here.

V. Summary

The yields of isotopes formed in the spallation of arsenic and the extrapolation of these yields to the neighboring isotopes has led to the following statements.

1) The yields of arsenic isotopes are higher than those of selenium isotopes because the former originate from non-capture excitation processes.

2) The large yields of isotopes from arsenic to zinc are due to non-capture excitation processes for the isotopes near arsenic.

Progressing through zinc to lower elements a larger fraction of the yield is due to capture processes.

3) The lowest isotopes observed, in the region of manganese and down to chlorine are formed from capture processes in which chiefly alphas are emitted.

4) Since the formation of Cl^{38} requires 168 Mev, 8 alpha particles must be emitted with only slightly more energy than the barrier height.

5) A high energy deexcitation occurs by successive emissions of neutrons, protons, and alpha particles. The course of the reaction follows the line of most probable formation, which below copper follows the valley of the energy surface.

6) The yields of isobars increase smoothly towards a curve running from arsenic one or two units above the line of stability at masses 70 through 67, then approaching the line of stability.

7) The yields of isotopes of given Z increase smoothly towards this line of most probable formation.

8) The total yield of isobar A plotted against A is a smoothly decreasing function. There is an unexplained hump at masses 54, 55 and 56.

9) The total yield of isotopes of given Z decreases with Z. The value for Z = 24 is high.

10) If we consider spallation reactions which would appear as nuclear stars, the mean prong number is 1.6 less than that predicted from nuclear star theory.

11) There are no pronounced differences with the yield patterns of spallation products from copper and antimony.

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Fig. I

Diagram of Separation Procedures.

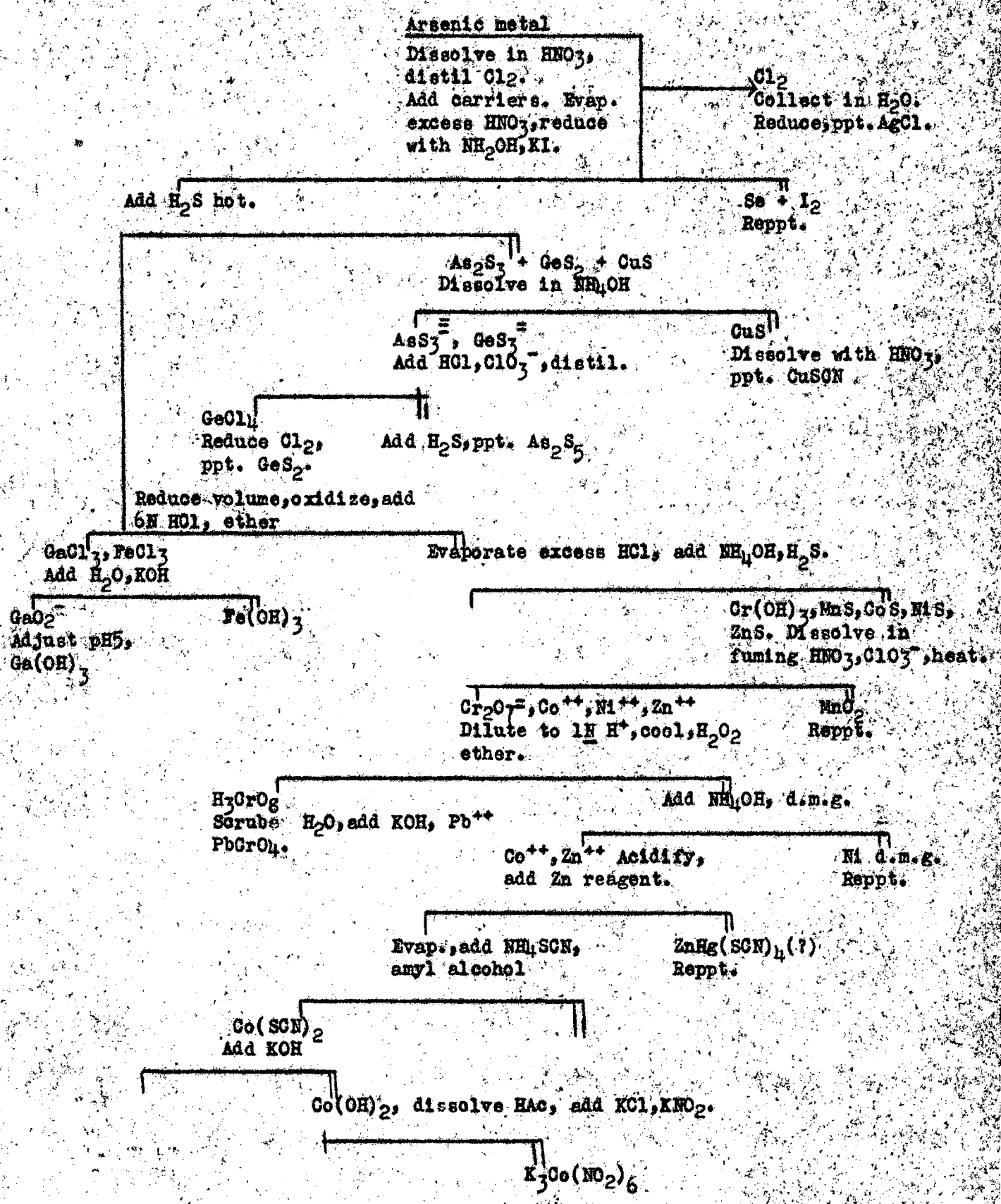

FIG. II

EXPERIMENTAL YIELDS $_{33}As^{75} + D^{+}$ (190 MEV)

RELATIVE TO As^{72}

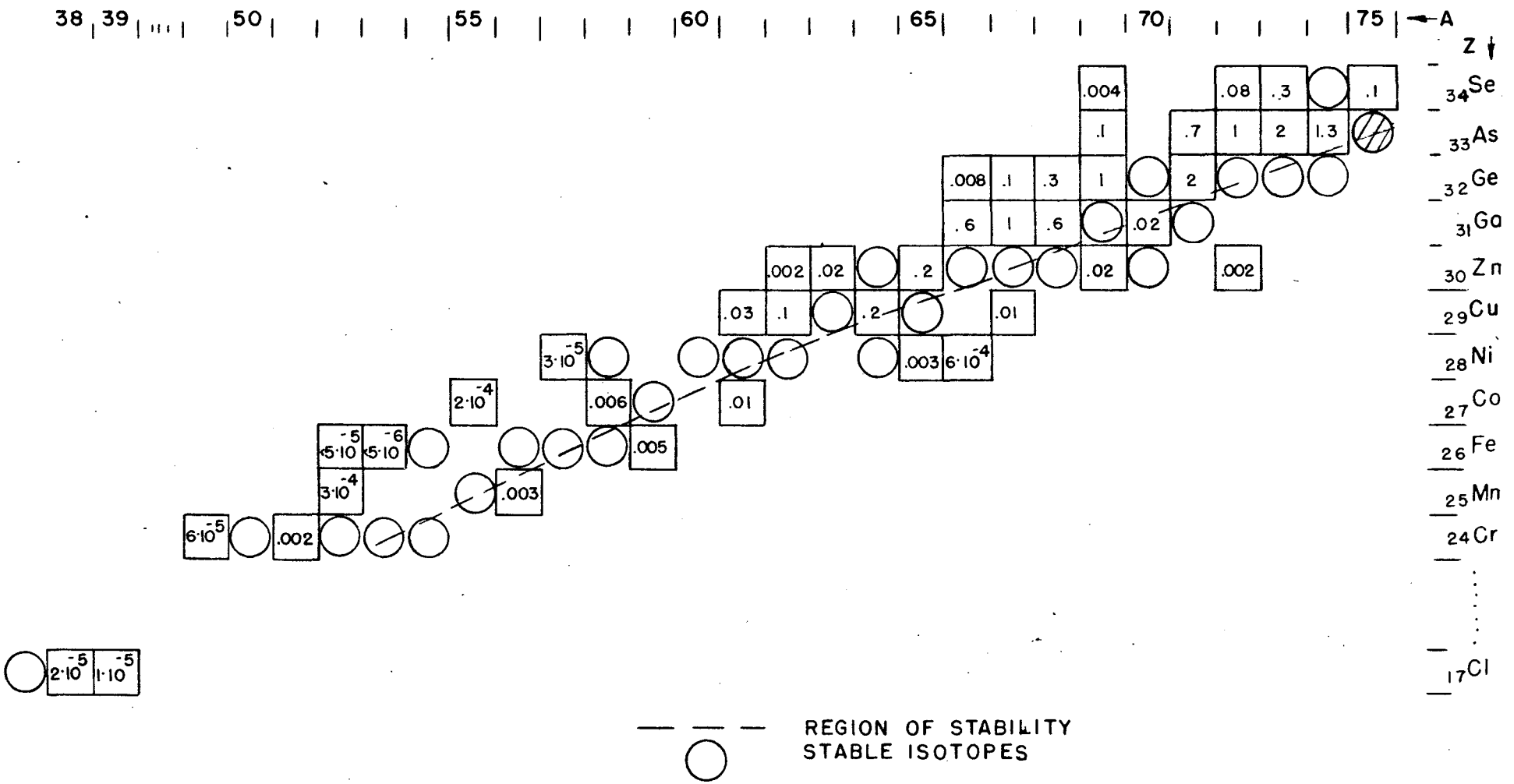

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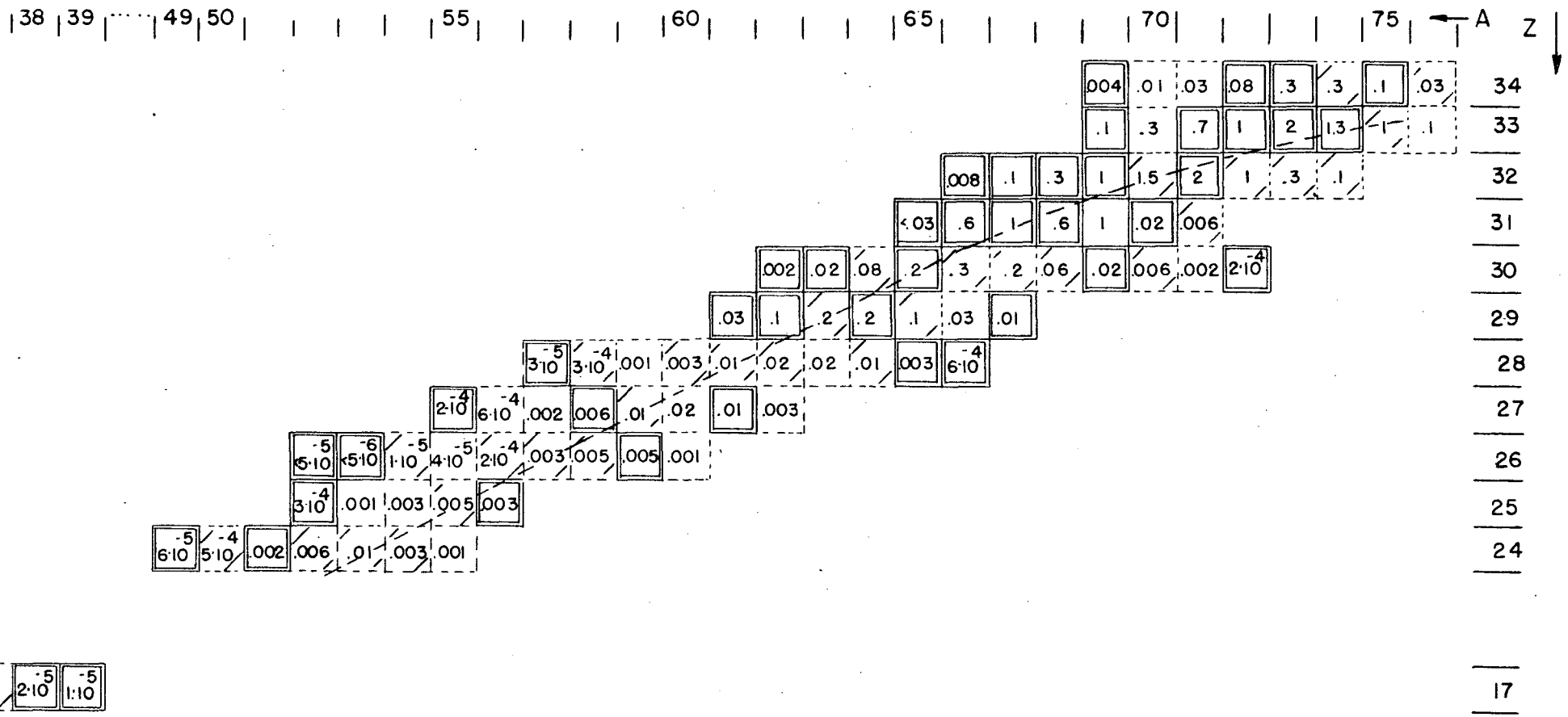
FIG. III

RELATIVE YIELD DISTRIBUTION [EXPERIMENTAL AND INTERPOLATED]



YIELDS RELATIVE TO As^{72}

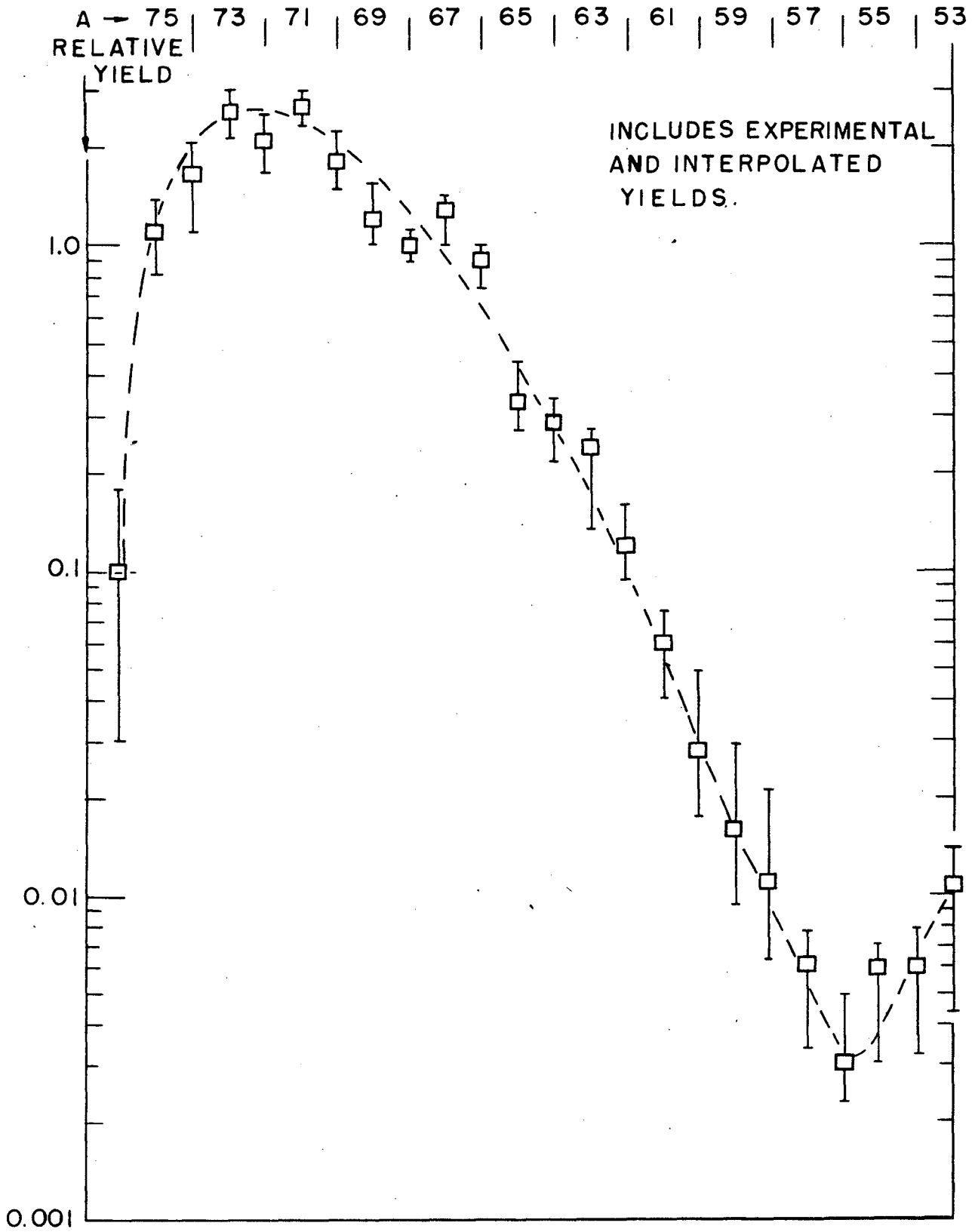
INTERPOLATED YIELDS

STABLE ISOTOPES

FIG. IV

YIELD MASS NUMBER DISTRIBUTION

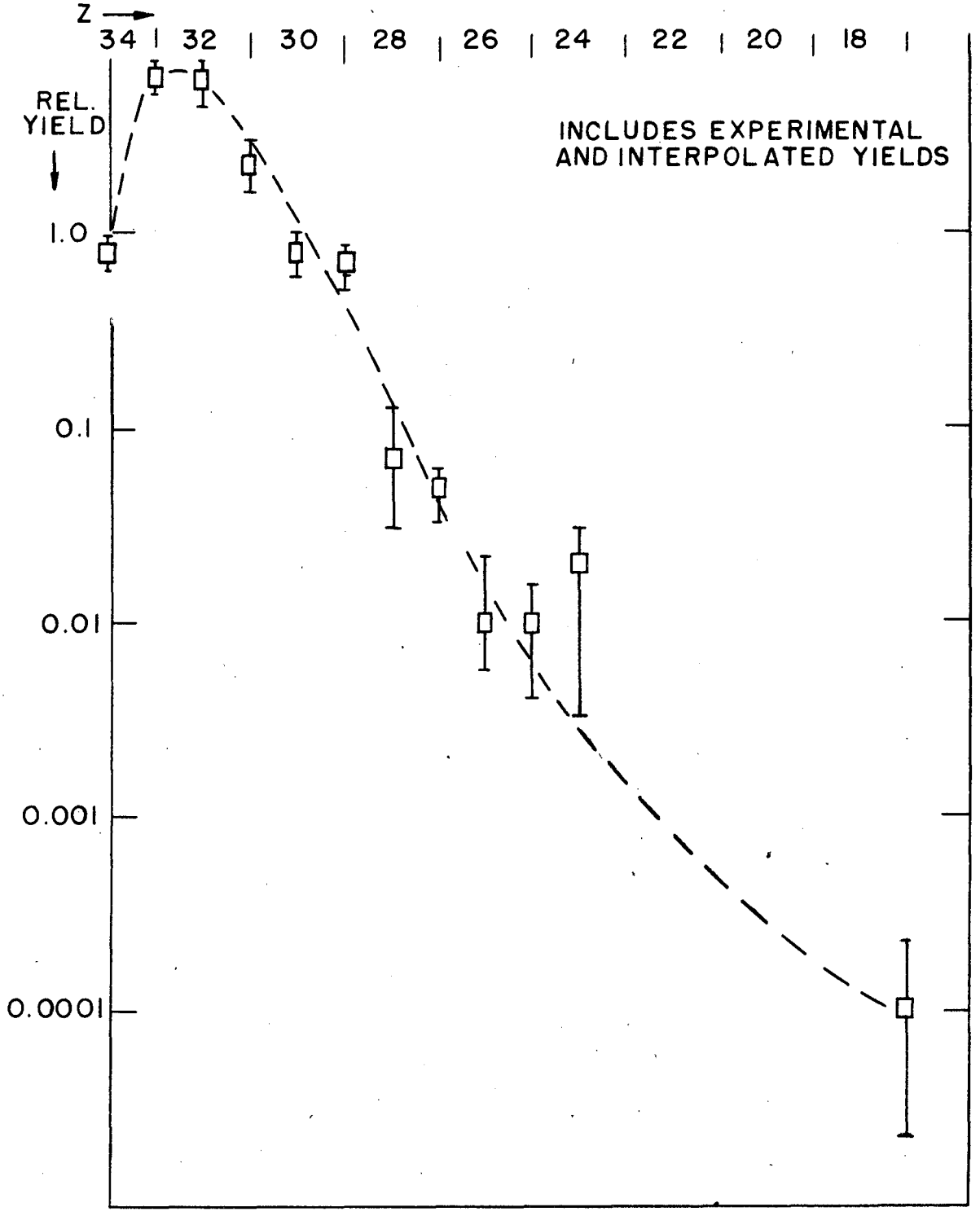
TOTAL YIELD OF GIVEN A RELATIVE TO As^{72}



13516 -1

FIG. V

YIELD DISTRIBUTION AMONG THE ELEMENTS
RELATIVE TO As^{72}



13517-1

FIG. VI

APPROXIMATE MINIMUM EXCITATION ENERGIES $As^{75} \longrightarrow Z^{A}$

Z \ A	38	39	50	55	60	65	70	75
33							45	36	27	18	9	0
32							76	67	58	49	40	31	22	13
31							65	56	47	38	29	20	35	26
30					103	94	85	76	66	58	51	42	33	48	39
29					87	78	69	59	51	41	32
28			125	116	103	90	85	73	64	55	54	45
27				118	109	100	91	82	73	64
26			147	138	129	119	109	97	86	77
25			122	113	104	95	86
24		150	141	132	123	110	99	88
⋮															
17	168	159													

CALCULATED MASS OF As^{75} = 74.947

VALUES CALCULATED FROM EXACT MASSES (9)

VALUES ARE FOR EMISSION OF THE MAXIMUM NUMBER OF ALPHAS