Excitation Functions for Spallation Reactions on Cu*

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Excitation functions have been measured for 190-Mev deuterons from the 184-inch Berkeley cyclotron on the natural isotopic mixture of Cu^{63} and Cu^{65}. Chemical separation of Zn, Cu, Ni, Co, Fe, and Mn has been performed and the yields of several of the radioactive isotopes of these elements have been measured as a function of deuteron energy. The results seem to be in agreement with Serber's theory of high energy processes and with other information on the yields of nuclear reactions at these energies.

I. INTRODUCTION

MEASUREMENTS of the yields of nuclear reactions as a function of the energy of the bombarding particles have provided considerable information regarding the mechanics of the nuclear processes involved. At low energies the familiar type of excitation function is one which rises from the threshold to a maximum and then falls off rapidly owing to the competition from other processes. Most of these results are explained satisfactorily by the theory of the compound nucleus formed by the addition of the bombarding particle. Early results with the very high energy particles from the 184-inch cyclotron showed a considerably different behavior in that the yield did not fall off at high energies. Serber[1] has explained these results in terms of the inelastic scattering of the impinging particle leaving in turn an excited nucleus. This excited nucleus is now the compound nucleus and decays by the usual processes. More detailed calculations have been made for the case of $C^{12}(p, pn)O^{11}$ by Heckrotte and Wolff.[2]

II. METHOD

This work was undertaken for the purpose of making a thorough study of the different radioactivities resulting from the deuteron bombardment of a single element, copper. Lindner and Perlman[3] have discussed the factors involved in the spallation reactions from As. These factors will not be reviewed here. Excitation functions have been determined for Zn^{63}, Zn^{62}, Cu^{64}, Cu^{61}, Ni^{65}, Ni^{57}, Co^{58}, Co^{56}, Co^{55}, Fe^{59}, Fe^{52}, Mn^{56}, and Mn^{52}, formed by 194-Mev deuterons on natural isotopic copper. The stacked foil technique, described for the 184-inch cyclotron by Thornton and Senseman,[4] was used. The foils on which the chemical separations were performed were electrolytic copper 0.002 in. thick, 1-in. diameter disks. These were placed in a 2-in. square stack of $\frac{1}{32}$-in. copper absorbers so as to cover the energy ranges desired. It was not practicable to perform the separations on more than 12 foils, so 10 were placed between 194- and 0-Mev energies of the deuterons and 2 beyond the range to give an estimate of the neutron background and of any effect of deuterons scattered into the side of the stack. A 0.001-in. Al foil of the same size as the Cu foils was placed at the high energy edge of the stack and used as a monitor. The $Al^{27}(d, \alpha p)Na^{24}$ cross section of 0.048 barn[5] at 194 Mev was used to estimate the cross sections for the elements formed.

The stack of foils was bombarded in the internally deflected beam of the 184-inch cyclotron for times of the order of one hour. After chemical separations to be described below, the samples were counted with mica end window counters, and the resultant decay curves resolved to determine amounts of different isotopes present. No special attempt was made to determine isotopes with half-lives less than about one hour. The Al monitor was counted under the same geometry, as nearly as possible, in order to minimize effects of backscattering, etc.; that is, if the samples were plated on Pt disks, the monitor was mounted on such a disk, etc. Experiments showed variations in counting rates of as much as 100 percent, depending on the backing and geometry of the sample, so that the estimates of cross section are certainly no more accurate than a factor of 2. Fortunately, the 1.4-Mev β-spectrum of Na^{24} is a reasonable mean of those investigated, though this isotope was chosen because of the convenient half-life

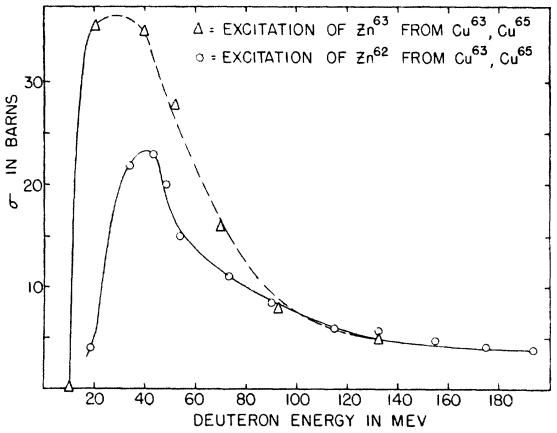

FIG. 1. Excitation functions of Zn^{63} and Zn^{62}.

* This work was performed under the auspices of the AEC.

[1] R. Serber, Phys. Rev. **72**, 1114 (1947).

[2] W. Heckrotte and P. Wolff, Phys. Rev. **73**, 264 (1948).

[3] M. Lindner and I. Perlman, Phys. Rev. **78**, 499 (1950).

[4] R. L. Thornton and R. W. Senseman, Phys. Rev. **72**, 872 (1947).

[5] H. W. Hubbard, Phys. Rev. **75**, 1470 (1949).

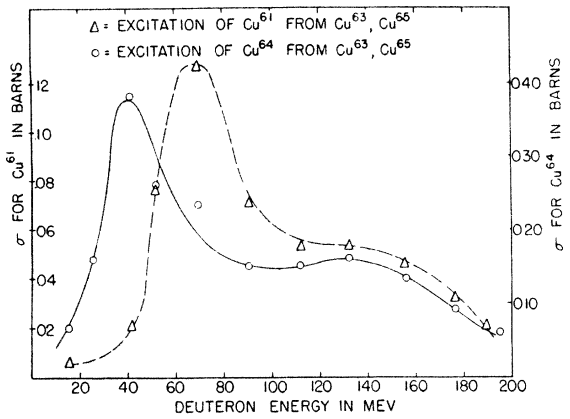

FIG. 2. Excitation function of Cu^{64} and Cu^{61}.

and absence of other activities. The excitation curve for the reaction $Al^{27}(d, \alpha p)Na^{24}$ was also used as a check on the energy values. It has a peak at approximately 25 Mev, which was the energy at the last Cu foil. The deuterons have passed through 22 g/cm² and the energy straggling is so large that no detail is possible below this.

Experimentally, the major problem was that of getting quantitative chemical separation of the elements near Cu from the 400 mg of Cu in the foils, and using at maximum about 3 mg of carrier. More carrier would have affected β-counting accuracy. The problem was met by special method of separation and duplicate equipment for processes such as ether extraction, etc., so that work on 12 foils could be completed in a few hours. The details are given in the appendix.

III. DISCUSSION OF RESULTS[6]

A. Zn^{63} and Zn^{62}

Zn^{63} is the well-known 38-min. β^+ activity formed in this case by $(d, 2n)$ and $(d, 4n)$ reactions; Zn^{62}, formed by $(d, 3n)$ and $(d, 5n)$, decays 90 percent by K-capture, 10 percent by β^+ emission, and is mainly detected by its daughter,[7] the 10-min Cu^{62}. The yields are shown on Fig. 1. These activities have the advantage that there is no background caused by neutrons, since Zn can be formed only by the addition of a charge. Each curve is presumably a composite of two double humped ones caused by the two reactions, but the energy resolution is not sufficient to show this up. The Zn^{62} curve is displaced to higher energy as would be expected. The fall off at high energies is not as rapid as might be expected from simple compound nucleus theory. This may be the result of exchange collisions between high energy incident protons and neutrons in the nucleus. The cross section at the maximum, 0.4 barn, is also of the expected order of magnitude, the geometrical cross section of a Cu nucleus being about 1 barn.

B. Cu^{64} and Cu^{61}

Cu^{64} is the familiar 12.8-hr. activity whose excitation curve by the reaction (d, p) has been studied by Livingston and Wright[8] and has a maximum below 10 Mev. Clearly, this portion of the curve is not visible in Fig. 2, where the yield of Cu^{64} is plotted. (Note the different ordinates for Cu^{64} and Cu^{61}.) The maximum at approximately 40 Mev is presumably due to the reaction (d, dn), $(d, p2n)$, or $(p, pn)(n, 2n)$ depending on whether one considers the reaction as caused by the deuteron as a whole or by one of its constituents. One might expect the maximum of these reactions to be at about twice the threshold energy[2] or about 20 Mev.

Cu^{61} (Fig. 2) is the well known 3.4-hr. activity. It requires the expulsion of more neutrons and, consequently, the maximum in its curve is at higher energy than that for Cu^{64}. The shape suggests a combination of sharper resonance with slowly varying portion. Why this second portion should decrease above 140 Mev is not at all clear, however. None of the curves calculated by Heckrotte and Wolff show such a decrease, even for the case of C, a much more transparent nucleus. The maximum cross section for the production of Cu^{61} is also smaller than that for Cu^{64}, as would be expected from the additional competition at higher energies.

C. Ni

The two Ni isotopes measured were Ni^{65} and Ni^{57}. The circled points on Fig. 3 were adjusted to fit a smooth Ni^{65} curve. When so adjusted, the Ni^{57} points fit on a smooth curve. This seems to be good evidence that there was chemical loss in these samples. The magnitude of the corrections was about 20 percent. Ni^{65} is formed by the $(d, 2p)$ or (n, p) reactions, and so is present even beyond the range of the deuterons owing to neutrons formed in the stack. The threshold for the reaction is low (2.3 Mev), and since the strag-

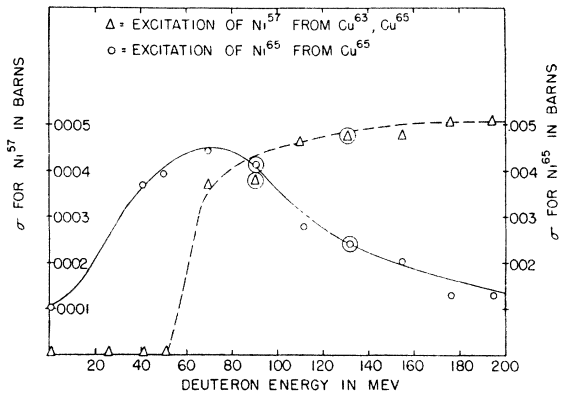

FIG. 3. Excitation functions of Ni^{57} and Ni^{65}.

[6] The periods, radiations, etc., of the radioactive isotopes are given in the table of Seaborg and Perlman, Rev. Mod. Phys. **20**, 585 (1948).

[7] R. W. Hayward, Phys. Rev. **79**, 541 (1950).

[8] R. S. Livingston and B. T. Wright, Phys. Rev. **58**, 656 (1940).

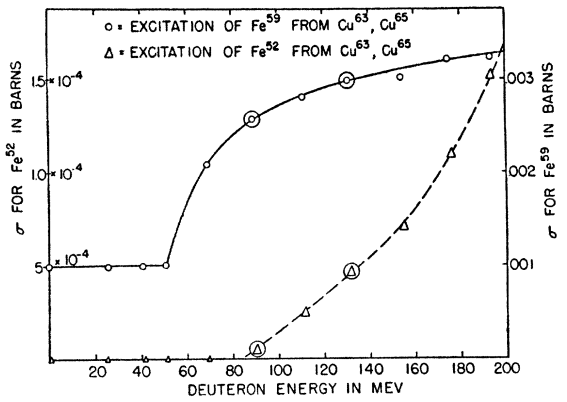

FIG. 4. Excitation functions of Fe^{59} and Fe^{52}.

gling is much greater than 2 Mev, a yield should be observed down to zero energy. The rather broad apparent maximum in the curve at about 60 Mev probably occurs at a somewhat lower energy. This energy is undoubtedly associated with the necessity for ejecting one or two protons over the Coulomb barrier. If the stripping process, in which the neutron enters the nucleus and the proton goes on, gives a large contribution, the necessary deuteron energy will be high because the neutron averages only half of it. In this connection, the yield of the (n, p) reaction as a function of neutron energy would be valuable to know.

The Ni^{57} curve is characteristic of those with high threshold energies. If one calculates the threshold for $Cu^{63}(d, 2p6n)Ni^{57}$ from a semi-empirical mass formula,[9] one gets 62 Mev. The agreement with the curve can be considered satisfactory. As will be seen below, there is evidence that α-particles are ejected as units; but in this case, unless the deuteron is captured to form a Zn compound nucleus, α-ejections cannot give Ni^{57}.

It is probably significant that the decrease of the Ni^{65}, Zn^{62}, and Cu^{61} cross sections sets in at about 60 Mev, when the Ni^{57} and others listed below start to rise.

D. Co

The chemical recoveries on Co were the least satisfactory of all those performed. The activities recovered were those of Co^{58} (72 days), Co^{56} (72 days), and Co^{55} (18 hours). In order to separate the first two, which have the same half-life, the particles were counted in a rough 180° magnetic spectrograph. The Co^{56} with a a 1.5-Mev beta-spectrum could then be distinguished from the 0.47-Mev spectrum of Co^{58}. A pure standard of the latter activity was also prepared by bombarding Mn with low energy alphas. Since Co^{58} decays only 15 percent by positron emission, there is a rather large correction to the observed counting rate; but the measurement in the magnetic spectrograph made certain that none of the x-rays were counted.

The excitation curves are of the type which rise to a fairly constant yield between 100 and 200 Mev. The surprising thing about these curves, however, is the large yield, 0.2 barn in the case of Co^{58} and 0.04 barn in the case of Co^{56} and Co^{55}. The results of Good, Peaslee, and Deutsch[10] on the positron K-capture branching should be accurate, so that there is no reason to suspect an error in this measurement of the large cross section of Co^{58}. It is to be noted that the reactions yielding Co^{58} are ones which might be written $(d, d\alpha n)$ and $(d, d\alpha 3n)$. The unusually high cross section for those reactions in which alpha-particles can be emitted after the excitation of the bombarded nucleus is discussed below.

Co^{57} (270 d.) is undoubtedly formed also in these experiments. Since these activities were weak, it was not possible to detect Co^{57} with certainty. However, the yields of Co^{56} and Co^{58} may be somewhat too high.

E. Fe

The 7.8-hr. Fe^{52}, which is the parent of the 21-m. Mn^{52m}, and the 49-d. Fe^{59} were the iron activities investigated. The results are shown in Fig. 4. Corrections similar to those mentioned for Ni were made on the circled points of the Fe^{59} curve with the resultant good fit to the Fe^{52} curve.

The Fe^{59} yield extends apparently to zero energy; and, while the energy definition is poor in this region, the curve does indicate that the reaction proceeds at low energies. Using the semi-empirical mass formula, one finds that the reaction $Cu^{65}(d, 2\alpha)Fe^{59}$ is exoenergetic by about 2 Mev. At low energies the spread in deuteron beam energy is large compared to the α-barrier height, so that one expects to find a yield at zero energy. The threshold for $Cu^{63}(d, 2p\alpha)Fe^{59}$ is 5 Mev, and because of the barrier for the α-particle this reaction will not become appreciable until higher energies. The rise in the curve beginning at about 50 Mev might

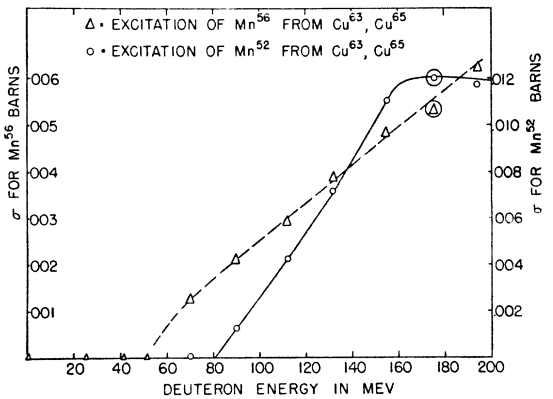

FIG. 5. Excitation functions of Mn^{56} and Mn^{52}.

[9] See Orear, Rosenfeld, and Schluter, "Nuclear Physics, Lectures by E. Fermi," p. 6. No Coulomb barrier energies are included in the threshold calculations.

[10] Good, Peaslee, and Deutsch, Phys. Rev. 69, 313 (1946).

e associated with this reaction or with reactions such s $Cu^{63}(n, p\alpha)$ or $Cu^{63}(n, 3p2n)$. In the latter the threshold is 28 Mev higher, while in both the necessary deuteron energy is increased because the neutron is considered as the particle involved and averages only one-half the deuteron energy. Further work on these reactions using low energy deuterons is projected.

The Fe^{52} exhibits the lowest cross section and the highest threshold measured in this work. The calculated threshold for $Cu^{63}(d, 4p9n)Fe^{52}$ is 104 Mev. The energetic threshold seems to be definitely below this (the energy definition in this region should be good) and would indicate that a reaction such as $Cu^{63}(d, \alpha 2p7n)$ or $Cu^{63}(n, \alpha p7n)$ was participating. Such reactions are particularly necessary if one postulates that only the neutron or the proton gives energy to the nucleus, although it is important to remember that this cross section is very small, and that occasionally a proton or a neutron may impart a large fraction of the deuteron energy to a nucleus.[11]

F. Mn

Mn^{56} (2.56 hr.) and Mn^{52} (6.5 d.) were the two Mn isotopes investigated. The curves are shown in Fig. 5. The 6.5-day activity is the lower of the two Mn^{52} isomeric states and is undoubtedly produced to some extent as a decay product of Fe^{52} through Mn^{52m}. The separations of Fe and Mn were performed on all samples at the same time and within a few hours of the end of the bombardment; and since the excitation curves are not radically different, the curve shown in Fig. 5 should not be much in error. Both Mn curves show the rather steep rise beginning at high energies characteristic of a reaction with high threshold. The calculated threshold for protons and neutrons out $(d, 5p4n)$ for Mn^{56} is 67 Mev, which is slightly above the observed threshold, indicating that probably α's are involved, while the calculated threshold for Mn^{52} from Cu^{63} is 102 Mev, and the observed threshold is definitely lower. In this case it seems certain that α's rather than individual protons and neutrons are emitted.

IV. CONCLUSIONS

The sets of yield curves presented above definitely show that at high energies of the bombarding particle the typical curve predicted by the compound nucleus theory at low energies is not found. The curves are in agreement with what would be expected from the theory presented by Serber. Detailed calculations such as extensions of those of Horning and Baumhoff[12] might be worth while to show whether the agreement is as close as could be expected.

More striking is the alternation of the magnitude of cross sections with the Z of the product isotope superimposed on a decreasing trend. Table I exemplifies this at 190 Mev. Since one cannot measure the yield of every isotope, the data are only fragmentary; but it does seem that the yield of Cu is large, of Ni small, of Co large, etc. A good example seems to be the cases of Fe^{52} and Mn^{52}, in which the latter cross section is a factor of 100 higher although it requires the ejection of an extra proton rather than a neutron. At very high energies one might expect that any barrier effect would be negligible and that the yields would be about equal. If we assume that the incident particle, neutron, proton, or deuteron, does not stick but only leaves the struck nucleus excited, then we notice that Mn^{52} can be formed by the ejection of two alphas and three neutrons from Cu^{63}, while Fe^{52} must be formed by ejection of particles less tightly bound to each other. As was mentioned above, the thresholds seem to indicate that alphas may be emitted; and this alternation in yield seems to be more evidence for the same thing. Such an alternation is evident in the results of Miller, Thompson, and Cunningham[13] with Cu and also, though not so noticeably, of Hopkins and Cunningham[14] working with As.

TABLE I. Maximum cross sections in barns for spallation products from deuterons on Cu. All are for 190 Mev except Cu^{61}.

A ↓ \ Z →	Mn	Fe	Co	Ni	Cu
61					0.11
60					
59		0.003			
58			0.200		
57				0.0005	
56	0.006		0.040		
55			0.040		
52	0.012	0.00015			

APPENDIX

The following chemical separations were mostly determined by convenience and the relative amounts of the activities present. The Zn separation had to be good only for Cu removal, since relative amounts of the other activities were small. The Ni, Co, Mn, and Fe separations had to be good from all other activities. The Fe separation was the most rigorous of them all, having to be good to one part in 10^6 from all other elements.

Zinc

The separation of Zn from the elements near it was as follows: Cu target foils were dissolved in the minimum necessary 6N HNO_3 and 2 mg of Zn carrier added as the nitrate or sulfate. One ml of concentrated H_2SO_4 was added and the HNO_3 removed by evaporating to SO_3 fumes. The solution was then diluted to about 3N H_2SO_4 and 500 mg of 20 mesh Al added. This solution was boiled until all the Cu was reduced and for about 5 minutes after the solution appeared water white. The solution was then cooled and filtered through a rapid filter paper, the Cu metal being washed with 0.1N H_2SO_4. The pH was adjusted to about 2 or 3 with NaOH and dilute H_2SO_4. The solution was then saturated

[11] R. Serber, Phys. Rev. **72**, 1008 (1947).
[12] W. Horning and L. Baumhoff, Phys. Rev. **75**, 370 (1949).
[13] Miller, Thompson, and Cunningham, Phys. Rev. **74**, 347 (1948).
[14] H. H. Hopkins and B. B. Cunningham, Phys. Rev. **73**, 1406 (1948).

with H_2S to precipitate ZnS, centrifuged, and the supernatant, which contained Mn, Fe, Co, and Ni, decanted. The precipitate was washed with 0.01N H_2SO_4 containing H_2S and then dissolved in 3 ml concentrated HNO_3. This solution was then evaporated to dryness on a stainless steel dish, flamed to destroy any excess sulfur and water of hydration, and was ready to be counted.

Copper

The Cu target foil was dissolved in a minimum of dilute HNO_3, made up to 250 ml in a volumetric flask and some convenient small fraction (0.8 percent to 4 percent) transferred to an electrolytic cell. Two drops of concentrated H_2SO_4 and a small amount of dilute HNO_3 were added and the solution plated to exhaustion by about ½ to 1 hour plating at about 20 milliamperes out of a volume of 20 ml. Zn, Mn, Fe, Co, and Ni will not plate under these conditions. The plates were quickly removed, washed in alcohol, dried in air, and were ready to count. The Cu was about 2 mg/cm² thick.

Nickel

The separation of Ni from the elements in the region of copper was done as follows: The target foil was dissolved in 6N HCl and a few drops of 30-percent H_2O_2, the excess H_2O_2 then being boiled off. About one-half to one mg of Ni carrier was added as chloride, the solution neutralized with NH_4OH, and then made slightly acid with HCl. NH_4HSO_3 was added to reduce Cu^{++} to Cu^{+} and excess SO_2 boiled off. CuSCN was precipitated from the warm solution with a few crystals of NH_4SCN, and allowed to settle for ten minutes. The solution was filtered and the precipitate washed with a one-percent solution of NH_4SCN containing a little NH_4SO_3. To complex the Fe, one ml of 50-percent tarataric acid was added to the filtrate which was made slightly ammoniacal and brought to near boiling. The Co, Zn, and Cu were thus complexed in the NH_4OH and Ni dimethyl glyoxime was precipitated by adding one to two ml of 1-percent dimethyl-glyoxime in ethanol. The precipitate was filtered and washed with hot water, dissolved in dilute HCl, and reprecipitated twice in order to free it from occluded Zn. Finally, the clean precipitate was dissolved in concentrated HNO_3, evaporated to dryness in a stainless steel dish, flamed, and was ready to count.

Cobalt

The separation of Co from the elements near Cu presented more difficulty than most of the other separations. The method finally evolved essentially consisted of removing all contaminant elements which would plate electrolytically and then plating the remaining Co onto platinum disks out of ammoniacal solution.

The Cu target foil was dissolved in minimum amount of dilute HNO_3, 1 mg each of Co, Ni, and Mn carriers, 2 drops of concentrated H_2SO_4 were added, and the solution diluted to 20 ml. The excess Cu was removed by electrodeposition on a stainless steel strip cathode set into the beaker using about 1 to 2 amp. at 2 to 3 volts. When the solution was water white, the electrodes were removed, 1 ml concentrated H_2SO_4 added and the solution evaporated to SO_3 fumes to destroy HNO_3 present. Then it was diluted to 35 to 50 ml and saturated with H_2S to precipitate the remaining Cu. Most of the Zn occluded on these CuS precipitates. After repetition of the procedure, the filtrate was again boiled to expel H_2S, made slightly ammoniacal, and 1 to 2 ml of 1-percent dimethyl-glyoxime in ethanol added. The Ni and Fe precipitates were filtered off, the filtrate was acidified with HNO_3 and evaporated to SO_3 fumes to destroy the alcohol, a few drops of concentrated HCl were added, and the filtrate was again taken to SO_3 fumes to insure removal of NO_3. The solution was transferred to an electrolysis cell fitted with a Pt disk cathode, made strongly ammoniacal, and Co metal deposited as a smooth, uniform, adherent plate on the cathode by plating for from ½ to 1 hour at about 1 to 2 amperes. MnO_2 was deposited simultaneously on the stirrer anode. Zn does not plate at these voltages. The electrolyte was then removed and replaced by distilled water with the current still on in order to prevent the Co plate from dissolving. The anode was quickly removed and the Co plate washed in alcohol and dried in air.

Manganese

It was found that Mn could be separated as part of the process for Fe isolation. The foils were dissolved in a minimum of 6N HNO_3 and 1 mg Mn carrier was added as the nitrate. The solution was then diluted to about 1N HNO_3, 0.1 g of $KBrO_3$ was added, and the solution was boiled for ten minutes or until the MnO_2 was well coagulated. There was probably some occlusion of Cu, Co, and Zn on this precipitate. The precipitate was filtered and washed with 6N HNO_3 until no Cu blue showed in the wash, then redissolved through the filter paper with concentrated HCl, and and 2 mg Cu carrier added. The solution was made strongly ammoniacal and a few crystals of $(NH_4)_2S_2O_8$ added. Any occluded Cu, Co, and Zn was complexed in the NH_4OH. The excess persulfate was boiled off, and the MnO_2 reprecipitated and centrifuged. After washing with dilute NH_4OH, the precipitate was deposited on a stainless steel dish and dried. This separation was perhaps the least troublesome of all those done in that the time required was a minimum and the degree of purification and percentage recovery excellent.

Iron

The following separation for Fe has the advantages of high purity ($\sim 10^6$) and that a small amount of carrier is necessary. Since the Fe yield was very small, the Fe half-lives relatively long, and the Fe^{59} β^- soft, these advantages turned out to be absolutely essential, even if recovery was not as complete as in some of the other fractions.

Cu foils were dissolved in a minimum 6N HNO_3 and diluted to 1 or 2 N HNO_3. One mg Fe and 1 mg Mn carrier were added as nitrates. The Mn was precipitated as MnO_2 by addition of about 0.1 g $KBrO_3$ and boiling for 10 minutes, filtered and washed. $Fe(OH)_3$ was precipitated from the filtrate by addition of excess NH_4OH and centrifuged. The Cu, Co, Ni, and Zn complexed in the supernatant liquid. The $Fe(OH)_3$ was dissolved in HCl, about 3 mg of $CuCl_2$ added, and the Fe precipitated twice more, this number of sweeps being necessary to get rid of the tremendous excess of Cu activity. The final $Fe(OH)_3$ precipitate was dissolved in three drops of concentrated HCl, and transferred to a separatory funnel with 6N HCl. An equal volume of diethyl ether presaturated with 6N HCl was added and the Fe extracted. Extraction was repeated on the aqueous layer, 1 mg Fe carrier added to this aqueous layer, and extracted again. All ether phases were collected with water and the ether boiled off in a hot water bath, then $Fe(OH)_3$ precipitated from this solution. This was dissolved in several drops concentrated HNO_3, evaporated to dryness on a stainless steel dish, flamed, and was ready for counting.