Radioactive Isotopes of Iron

J. J. LIVINGOOD, *Radiation Laboratory, Department of Physics, University of California*

AND

G. T. SEABORG, *Department of Chemistry, University of California, Berkeley, California*

(Received May 10, 1938)

Radioactive Fe^{59} has been produced through the reactions $Fe^{58}(d, p)Fe^{59}$ and $Co^{59}(n, p)Fe^{59}$; the half-life is 47 ± 3 days. The emitted particles are negative electrons, most of which have a range of 0.09 grams/cm² Al, while a small number extend to 0.35 grams/cm² Al. The gamma-ray shows a half-thickness of 10 grams/cm² Pb. Radioactive Fe^{53} is formed by the two processes $Fe^{54}(n, 2n)Fe^{53}$ and $Cr^{50}(\alpha, n)Fe^{53}$; positrons are ejected with a half-life of 8.9 ± 0.2 minutes. No subsequent decay of Mn^{53} to Cr^{53} has been observed. No activity corresponding to Fe^{55} has been detected, although Cr, Mn, Fe, Co and Ni have been bombarded with protons, neutrons, deuterons and alpha-particles in all the combinations that might be expected to produce this isotope; it is concluded that Fe^{55} is either stable or has a very long or a very short life.

INTRODUCTION

EVIDENCE is here presented for the production of a new negative electron emitting radio isotope of 47 ± 3 days half-life ascribed to Fe^{59}, which is made by the reactions $Fe^{58}(d, p)Fe^{59}$ and $Co^{59}(n, p)Fe^{59}$, and for a positron emitter of 8.9 ± 0.2 minutes half-life assigned to Fe^{53}, produced by the process $Fe^{54}(n, 2n)Fe^{53}$, with confirmation of the alternative mode $Cr^{50}(\alpha, n)Fe^{53}$.

APPARATUS

Activations were made with the Berkeley cyclotron, with deuteron currents of 100 to 20 microamperes at 5 to 8 Mev, proton currents of 100 microamperes at 3.2 Mev and alpha-particle currents of 0.5 microampere at 16 Mev. Sheet iron of known purity from various sources was used; this was often simply screwed to the water-cooled target holder, but for the more energetic bombardments the periphery of the iron was directly soldered to a cooling pipe. C.P. oxides of manganese, cobalt and chromium were supported generally behind a 0.0005-inch foil of commercial aluminum; more recently (to avoid recoil impurities from the foil) these powders have been sprinkled onto a sloping corrugated block of carbon. The radioactivities were measured with a quartz fiber electroscope equipped with a window of 0.0001 inch aluminum. An activity of 1 division per second corresponds roughly to 1 microcurie. The background activity, equal to about 0.001 divisions per second, has been subtracted in all the data shown.

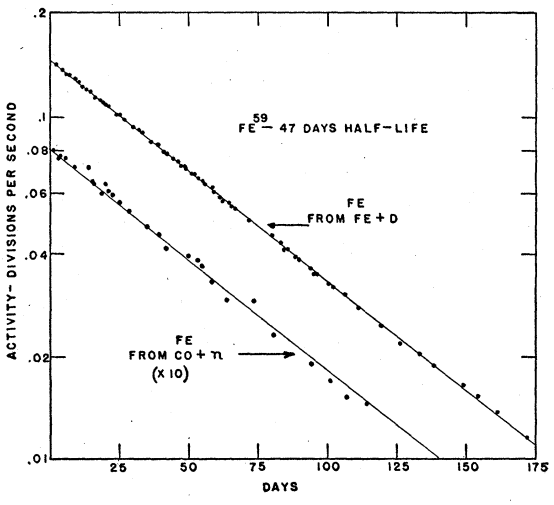

FIG. 1. Decay curves of Fe^{59}.

IRON ACTIVITY OF 47 DAYS HALF-LIFE FROM Fe+D

Figure 1 shows the decay curve of iron chemically separated from iron bombarded with 5.5 Mev deuterons for about 170 microampere hours. The half-life is 47 ± 3 days, this value superseding the 40-day figure given in a preliminary communication.[1]

The bombarded iron was dissolved in a mixture of HCl and HNO_3, and small amounts of $MnCl_2$

[1] Livingood, Seaborg and Fairbrother, Phys. Rev. **52**, 135 (1937).

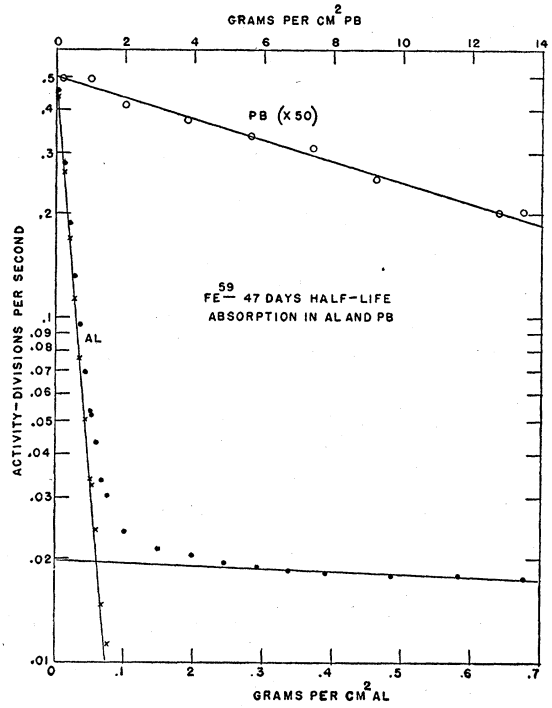

FIG. 2. Absorption curves of Fe^{59}.

and $CoCl_2$ were added to serve as carriers for the transmutation products. NaH_2PO_4 was also included to take care of the radio-sodium and radio-phosphorus generally present. The solution was made 6*N* in HCl and the iron was extracted as $FeCl_3$ by shaking with an equal volume of ether, according to the method of Noyes and Bray.[2] The ether containing the $FeCl_3$ was then always shaken with four or five successive portions of equal volumes of 6*N* HCl in order to remove all traces of radio Mn and Co. The complete absence of the very strong 18-hour Co and 2.6-hour Mn activities[1] in the iron fraction showed that this method offered a very good separation.

In some cases the $FeCl_3$ was converted into Fe_2O_3 after the separation, since it was found that Fe_2O_3 suffered fewer physical and chemical changes over a long period of time and so afforded better decay curves.

The iron was separated by an entirely different method on several occasions. $Fe(OH)_3$ was precipitated (after the removal of the manganese as MnO_2) by the addition of NH_4OH to a solution which contained a high concentration of ammonium ion. With this technique it was necessary to redissolve and reprecipitate the $Fe(OH)_3$ four or five times in order to effect a complete separation from cobalt. The iron fraction obtained in this manner showed the same 47-day activity as obtained in the ether extractions.

The absorption in aluminum of the 47-day activity is shown in Fig. 2. (These data were taken with a much stronger sample than that shown in Fig. 1.) The majority of the electrons have a range of about 0.09 grams/cm², while a weak tail extends out to 0.35 grams/cm². By the use of Feather's relation[3] ($E=R+0.091/0.511$ where the energy E is in Mev and the range R is in grams/cm²) the energies of the two groups are found to be 0.4 Mev and 0.9 Mev. With the range-energy relation of Widdowson and Champion[4] (wherein the two constants of Feather's equation are altered to 0.165 and 0.536) the energies are about 0.5 and 1.0 Mev.

The gamma-ray absorption is also shown in Fig. 2, indicating a half-thickness of 10 grams/cm² Pb, which corresponds to an energy of about 1 Mev from the data of Gentner.[5]

The sign of the particles has been observed to be negative, by the use of simple magnetic deflection through air and also by a cloud chamber with magnetic field and by a trochoid analyzer, for which we are indebted to Dr. F. N. D. Kurie and Dr. E. Lyman.

IRON ACTIVITY OF 47 DAYS HALF-LIFE FROM Co+*n*

Several grams of C.P. cobalt oxide were left for three months in close proximity to the target chamber of the cyclotron in order to receive the neutrons released from the bombardment with deuterons of beryllium or other targets. A chemical separation for manganese, iron and cobalt was made, and an activity of approximately 47 days half-life was found in the iron fraction, as previously reported.[1] It was not possible to check the sign of the particles in this

[2] Noyes and Bray, *Qualitative Analysis for the Rare Elements* (Macmillan Company, 1927).

[3] Feather, Phys. Rev. **35**, 1559 (1930).

[4] Widdowson and Champion, Proc. Phys. Soc. **50**, 192 (1938).

[5] Gentner, J. de phys. et rad. **6**, 274 (1935).

weak sample. The decay curve is shown in Fig. 1. That the activity is really due to a transmutation of cobalt and not to iron impurity in the cobalt oxide, is attested to by the fact that the 47-day period has not been obtained from direct bombardment of iron with neutrons (see below).

Identification of the 47 Day Activity with Fe^{59}

Figure 3 shows the percent abundance of the stable Cr, Mn, Fe, Co and Ni isotopes as quoted by Livingston and Bethe.[6] It is at once apparent that only Fe^{59} can be negative electron active. Furthermore, the only radio-iron that can be made from cobalt with neutrons is Fe^{59}, so that we are justified in ascribing the 47-day activity to this isotope. The two transmutations are

$$_{26}Fe^{58}+{}_1D^2\rightarrow{}_{26}Fe^{59}+{}_1H^1$$

and

$$_{27}Co^{59}+{}_0n^1\rightarrow{}_{26}Fe^{59}+{}_1H^1.$$

Each is followed by

$$_{26}Fe^{59}\rightarrow{}_{27}Co^{59}+{}_{-1}e^0 \quad (47\text{ days}).$$

It might reasonably be expected that this activity should also occur after neutron bombardment of iron through $Fe^{58}(n, \gamma)Fe^{59}$, but we have been unable to detect the effect, even when borings were taken from the iron pole pieces of the cyclotron which had received neutron bombardment for many months. This may be understood from the very small abundance of Fe^{58} (0.5 percent) and the relatively small number of neutrons compared to deuterons.

Andersen[7] has reported a 3-day activity, following neutron irradiation of cobalt, said to be chemically identified with iron. We have not

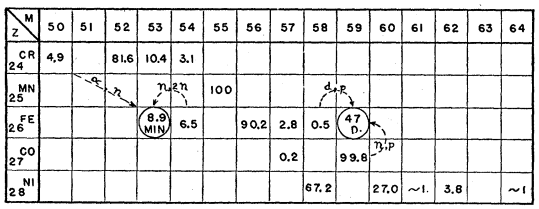

Fig. 3. Percent abundance of isotopes in the neighborhood of iron. Radioactive isotopes, with their half-lives, are encircled, the broken lines indicating the reactions by which they are produced.

[6] Livingston and Bethe, Rev. Mod. Phys. **9**, 380 (1938).
[7] Andersen, Nature **138**, 76 (1936).

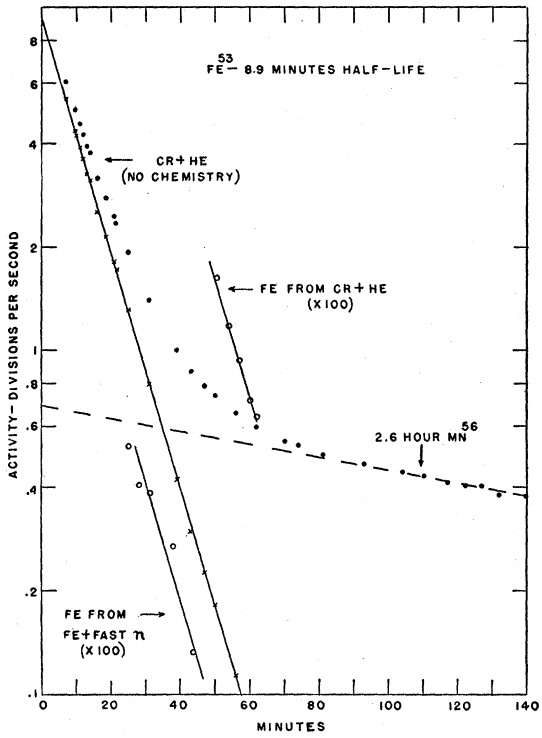

Fig. 4. Decay curves of Fe^{53}.

detected this period on any occasion and are inclined to believe it to represent a poor determination of the 47-day activity, probably because of the weakness of the source.

The bombardment of nickel with neutrons has thus far failed to produce the 47-day period, although the reaction $Ni^{62}(n, \alpha)Fe^{59}$ might be expected. Ni^{62} is present to only 3.8 percent, which may account for this failure.

Iron Activity of 9 Minutes Half-Life from Cr+He

Henderson and Ridenour[8] have reported a radioactive iron of 8.9 ± 0.3 minutes half-life, which emits positrons, and which is produced by bombarding chromium with electrically accelerated alpha-particles of 9 Mev energy. They also find an electron emitting manganese isotope of 2.66 ± 0.25 hours half-life as a result of the same bombardment and ascribe it to the well-known Mn^{56} isotope, according to the reaction $Cr^{53}(\alpha, p)Mn^{56}$.

[8] Henderson and Ridenour, Phys. Rev. **52**, 889 (1937).

We have repeated this experiment with 16 Mev alpha-particles and confirm their results in every essential particular. Our values for the half-lives are 8.9±0.2 minutes and 2.60±0.05 hours. The decay curves are shown in Fig. 4, both for chemically separated iron and for an untreated sample which shows both the iron and manganese periods.

Iron Activity of 9 Minutes Half-Life from Fe+Fast Neutrons

We have not observed any activities ascribable to iron when bombarding this metal with neutrons from Be+D, but when the fast neutrons from Li+D are used, the 9-minute decay is obtained. If no chemical separation is made, the activity is composite, the 9-minute period is superimposed on the 2.6 hour Mn^{56} period. The Mn^{56} is formed on this occasion by $Fe^{56}(n, p)Mn^{56}$. The decay of the separated iron is shown in Fig. 3.

Identification of the 9-Minute Activity with Fe^{53}

Henderson and Ridenour assigned the 9-minute positron emitting isotope to either Fe^{53} or Fe^{55} through the two alternatives $Cr^{50,52}(\alpha, n)Fe^{53,55}$. With the former choice, the difficulty arises as to the fate of the radio-Mn^{53} which must be formed by the decay of Fe^{53}, since no second period was observed. We also have been unable to find any evidence of the decay of Mn^{53}, but in spite of this we believe the 9-minute activity to be due to Fe^{53} rather than to Fe^{55} because:

(1) it is not produced by deuteron or slow neutron bombardment of Fe,

(2) it is produced by fast neutrons on Fe,

(3) attempts to produce Fe^{55} by other reactions have not disclosed a 9 minute activity (see below).

We therefore conclude that the 9-minute period is formed by

$$_{24}Cr^{50}+{}_{2}He^{4}\rightarrow{}_{26}Fe^{53}+{}_{0}n^{1}$$

and by $$_{26}Fe^{54}+{}_{0}n^{1}\rightarrow{}_{26}Fe^{53}+2{}_{0}n^{1},$$

followed by

$$_{26}Fe^{53}\rightarrow{}_{25}Mn^{53}+{}_{1}e^{0} \quad (8.9 \text{ minutes}),$$

while the decay of Mn^{53} to stable Cr^{53} is still undetected.

It is interesting to note that this fast neutron effect on iron has a half-life almost identical with the 8 to 10 minutes given by Clay and his associates when reporting[9,10] an activity induced in iron by cosmic rays. Recently Ramsey and the Montgomerys[11] have endeavored to repeat this cosmic-ray experiment, but were unable to confirm the effect.

Search for Fe^{55}

We have not found any iron activity after bombarding iron with protons at 3.6 Mev, although the transmutation $Mn^{55}(p, n)Fe^{55}$ might be expected. DuBridge and his associates have also made this experiment using 4 Mev protons and also report negative results.[12]

Only the reaction $Mn^{55}(d, 2n)Fe^{55}$ could lead to radioactive iron if manganese is exposed to deuterons. We have made this attempt on several occasions, but have not obtained any activities definitely attributable to iron.

The bombardment of chromium with alpha-particles could give $Cr^{52}(\alpha, n)Fe^{55}$, but the only iron period we have observed is the 9-minute one which has been assigned to Fe^{53} (see above).

Activation of cobalt with deuterons might be expected to produce Fe^{55} by the reaction $Co^{57}(d, \alpha)Fe^{55}$, but no iron activity has been found after a number of such attempts.

We have not found any iron periods after irradiating iron with slow neutrons and only the 9-minute period is disclosed when using fast neutrons. On one occasion the iron separated from a deuteron bombardment of iron was placed directly inside the electroscope, in case the radiations from Fe^{55} should not be able to penetrate the 0.0001-inch aluminum window; nothing but the 47-day period was observed.

Darling, Curtis and Cork[13] and ourselves[1] have found an 18-hour positron emitting cobalt isotope produced by deuterons on iron. This has been

[9] Clay and Van Tijn, Physica **4**, 909 (1937).
[10] Clay and Jonker, Physica **5**, 171 (1938).
[11] Ramsey, C. G. and D. D. Montgomery, Phys. Rev. **53**, 196 (1938).
[12] DuBridge, Barnes, Buck and Strain, Phys. Rev. **53**, 447 (1938).
[13] Darling, Curtis and Cork, Phys. Rev. **51**, 1011 (1937).

presumed to be Co^{55}, formed by $Fe^{54}(d, n)Co^{55}$. A second activity growing in the cobalt should be observed, if this is the case, as Co^{55} decays to Fe^{55} and this in turn to stable Mn^{55}. We have made very rapid separations of iron from radiocobalt when the 18-hour activity was exceedingly strong, hoping to detect a shorter lived iron, but nothing was found. This procedure has been repeated at a much later time, in case Fe^{55} had a considerably longer life which would require more time for it to grow to an observable amount, but again with negative results.

The existence of a positron emitting cobalt of several months half-life (formed from deuterons on iron) has been announced.[1] The possibility was considered that this isotope might be Co^{55} and hence might be the source of Fe^{55}. Iron has been separated from such cobalt samples of various ages, from a few days to several months, but again without disclosing any evidence for Fe^{55}.

We must therefore fall back on the usual conclusions in such cases: Fe^{55} is stable, or it has a very short or a very long lifetime. (We are continuing to follow some of our iron samples, in case an activity longer than 47 days should ultimately appear.)

Detection of Gallium Impurity in Iron

During the course of the deuteron bombardments of iron, there were repeatedly obtained in the ether extract of $FeCl_3$ not only the 47-day period but also weak activities of 18 to 25 minutes and 14 to 20 hours half-lives, both emitting negative electrons. These activities were so feeble as to make it extremely unlikely that they should be ascribed to iron. Therefore possible impurities in the iron were considered which could give these periods and whose chlorides would be extracted in the ether. $GaCl_3$ would be so extracted, and the periods of gallium under slow neutron bombardment have been given by Amaldi[14] as 20 minutes and 23 hours.

A chemical separation of gallium and iron was made after another deuteron bombardment of iron. A little gallium was added to the acid solution of activated iron, as well as the usual manganese and cobalt. Since $FeCl_2$ is not soluble in ether, the iron was reduced to this form by the addition of mercury, and the $GaCl_3$ was then extracted with ether from a solution $6N$ in HCl. The $FeCl_2$ was then oxidized to $FeCl_3$ with HNO_3, and the $FeCl_3$ was separated from the Mn and Co by extraction with ether. The iron fraction showed only the 47-day period, while the gallium exhibited periods of 23 minutes and 14 hours. In as much as this was not in total accord with the results of Amaldi, we felt it desirable to activate gallium with slow neutrons for an independent check. This was done and a chemical separation for gallium, zinc and copper was made. The gallium fraction showed half-lives of 23 minutes and 14 hours. These figures are in close agreement with the 20 minutes and 14.1 hours quoted by Sagane,[15] who activated gallium with slow neutrons but without benefit of chemical identification.

The relative intensities of these gallium and 47-day iron activities, together with the known isotopic abundances and the duration and intensity of the bombardments, allows an estimate to be made of the amount of gallium present in the iron, on the assumption that the cross section for capturing the neutron out of the deuteron is the same for gallium as for iron. For example, one iron sample showed a gallium content of 6 parts in 10^6. One tenth this much could have been detected readily.

A number of other weak activities, due to impurities, have been found in some of the other bombardments discussed above. In certain cases identification has been possible, while in others no positive assignment has as yet been made.

Acknowledgments

This research has been materially aided by the support of the Chemical Foundation, the Research Corporation and the Josiah Macy Jr. Foundation, to which we express our greatest appreciation.

[14] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti and Segrè, Proc. Roy. Soc. **A149**, 522 (1935).

[15] Sagane, Phys. Rev. **53**, 212 (1938).