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Radioactive Manganese Isotopes

J. J. LIVINGOOD, *Radiation Laboratory, Physics Department, University of California*
AND
G. T. SEABORG, *Chemistry Department, University of California, Berkeley, California*
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Five radioactive decay periods of manganese have been obtained by bombarding cobalt, iron, manganese, chromium and vanadium with neutrons, deuterons and alpha-particles. Arguments are given for the assignment to various isotopes, as follows:

REACTION	HALF-LIFE	REACTION	HALF-LIFE
$Cr^{50}(d,n)Mn^{51}$	46±2 min. (+)	$Mn^{55}(n,\gamma)Mn^{56}$	2.59±0.02 hours (−)
$Fe^{54}(d,\alpha)Mn^{52}$	21±2 min. (+)	$Mn^{55}(d,p)Mn^{56}$	
	6.5±1 days (+)	$Cr^{53}(\alpha,p)Mn^{56}$	
$Fe^{56}(d,\alpha)Mn^{54}$		$Fe^{58}(d,\alpha)Mn^{56}$	
$Cr^{53}(d,n)Mn^{54}$	310±20 days	$Fe^{58}(n,p)Mn^{56}$	
$V^{51}(\alpha,n)Mn^{54}$	(*k* electron capture)	$Co^{59}(n,\alpha)Mn^{56}$	

No evidence has been found for radioactive Mn^{53}. Chemical identification has been made in all instances, except for the case of the bombardment of vanadium with alpha-particles. The 46-minute period emits positrons with a range 0.9 gram/cm² Al and annihilation radiation absorbed to half-value by 5.0 grams/cm² Pb. The positrons from the 6.5-day activity have a range 0.2 gram/cm² Al. The 310-day activity consists of a soft radiation, whose absorption coefficient is consistent with that of the Cr $K\alpha$ x-ray line, and also a gamma-ray reduced to half-value by 8.4 grams/cm² Pb. The range of the electrons from the 2.6 hour Mn^{56} has been measured as 1.3±0.1 grams/cm² Al, while the gamma-ray is brought to half-value by 11.4 grams/cm² Pb.

INTRODUCTION

THE artificial radioactivities reported in this investigation were produced by the Berkeley cyclotron with deuterons at 5.5 Mev and 7.6 Mev and with helium ions at 16 Mev. The activities were observed with a quartz fiber electroscope. C.P. carbonates and oxides of manganese were supported in an envelope of thin aluminum foil; chromium was bombarded either as the oxide (similarly supported) or as the metal plated onto copper. Iron and vanadium were used in the metallic form. Short periods of the order of a few minutes were not looked for, in general.

The bombarded samples were dissolved in a mixture of HCl and HNO_3 and small amounts of the appropriate elements were added to act as carriers for the radioactive transmutation products. The solution was adjusted to a concentration of 16 *N* in HNO_3 for the precipitation of the manganese, which was brought down as MnO_2 by the addition of solid $KClO_3$ to the boiling solution. The MnO_2 was usually purified by redissolving it in an acid solution of H_2O_2 and by again precipitating from a boiling solution of 16 *N* HNO_3 by the addition of solid $KClO_3$.

Figure 1 shows the percent abundance of the stable isotopes in the region near manganese, as compiled by Livingston and Bethe.[1] The identity of the radioactive isotopes, shown in circles, is that derived from the present investigation.

[1] Livingston and Bethe, Rev. Mod. Phys. **9**, 380 (1937).

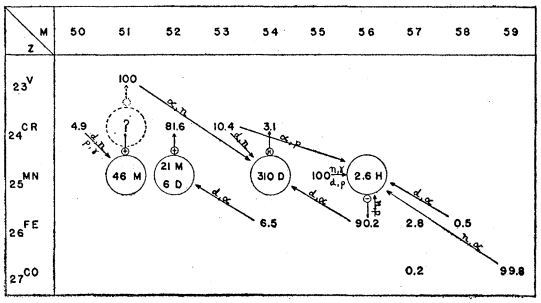

FIG. 1. Percent of stable isotopes in the neighborhood of Mn. The half-lives of radioactive isotopes are shown in circles, with arrows indicating known transmutations.

Mn^{51} FROM Cr+D AND Cr+H: HALF-LIFE 46±2MIN. (+)

We have briefly reported[2] the formation of a chemically identified manganese isotope with half-life 46 minutes that is formed by deuteron bombardment of chromium. Positrons are emitted. The Cr(d,n)Mn reaction could lead to Mn^{51}, Mn^{53} and Mn^{54}, and of these possibilities we believe the 46-minute activity must be assigned to Mn^{51}. If Mn^{54} were responsible, the activity should be apparent after deuteron bombardment of iron, through $Fe^{56}(d,\alpha)Mn^{54}$, and this is not found to occur; neither have we been able to produce this period by fast neutron bombardment of manganese, which would be expected to develop such a short-lived activity with considerable intensity, by the transmutation $Mn^{55}(n,2n)Mn^{54}$. Mn^{53} may be eliminated as the choice since radioactive Fe^{53}, with half-life 8.9 minutes, should then decay into Mn^{53} and ultimately show the 46-minute period. We have previously shown[3] that this does not occur, when Fe^{53} is produced by $Cr^{50}(\alpha,n)Fe^{53}$ and by $Fe^{54}(n,2n)Fe^{53}$. Ridenour and Henderson, who have also reported[4] the 8.9-minute iron activity after irradiating chromium with alpha-particles, were also unable to observe any second period corresponding to Mn^{53}.

The assignment of the 46-minute activity to Mn^{51} is consistent with the report by DuBridge[5] and his associates that proton bombardment of chromium gives rise to a positron emitting radioactivity of 42-minute period. Although chemical analysis was not made, this is doubtless the same isotope and the two modes of production are

$$_{24}Cr^{50}+{}_{1}D^{2}\rightarrow{}_{25}Mn^{51}+{}_{0}n^{1},$$
$$_{24}Cr^{50}+{}_{1}H^{1}\rightarrow{}_{25}Mn^{51}+\gamma$$

followed by

$$_{25}Mn^{51}\rightarrow{}_{24}Cr^{51}+{}_{+1}e^{0}\ (46\ \text{minutes}).$$

Neither DuBridge nor ourselves have detected a second activity corresponding to the decay of Cr^{51} to stable V^{51}; this indicates that Cr^{51} has a very short or a very long lifetime (or is possibly stable). This is consistent with the fact that the literature discloses no radioactive periods that are definitely due to chromium; in particular none ascribed to Cr^{51}.

The possibility that the 46-minute period could be due to Mn^{50} rather than to Mn^{51} cannot be excluded definitely, since the reactions $Cr^{50}(d,2n)Mn^{50}$ and $Cr^{50}(p,n)Mn^{50}$ might be the sources of the activity, which would then lead to stable Cr^{50}. A decision between Mn^{50} and Mn^{51} could be made by the bombardment of chromium with protons: a study of the excitation function for the 46-minute period would reveal whether the reaction $Cr^{50}(p,\gamma)Mn^{51}$ or $Cr^{50}(p,n)$

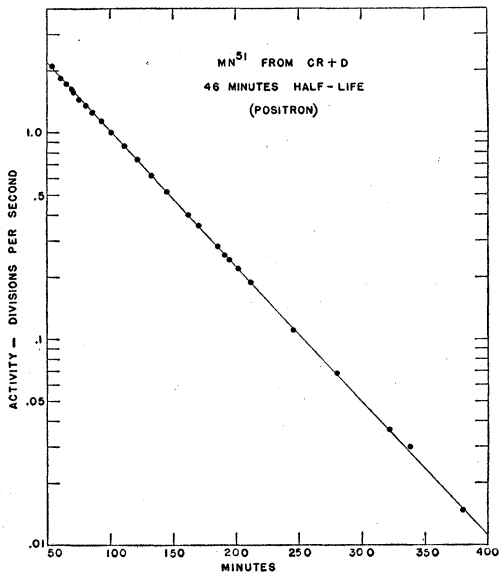

FIG. 2. Decay of Mn^{51}.

[2] Livingood, Seaborg and Fairbrother, Phys. Rev. **52**, 135 (1937).
[3] Livingood and Seaborg, Phys. Rev. **54**, 51 (1938).
[4] Ridenour and Henderson, Phys. Rev. **52**, 889 (1937).
[5] DuBridge, Barnes, Buck, Strain, Phys. Rev. **53**, 447 (1938).

Mn^{50} occurs, since the latter should exhibit a fairly sharp threshold.

Figure 2 shows the decay of manganese precipitated from chromium metal plated onto copper that was activated for 3 minutes with 5 microamperes of deuterons at 5.5 Mev. The half-life, as found from several determinations, may be given as 46 ± 2 minutes. Absorption curves are given in Fig. 3; the positrons have a range of 0.9 gram/cm² Al, unless modified by a line spectrum of electrons as pointed out below. This corresponds to a maximum energy of 2.0 Mev through the relation of Widdowson and Champion:[6]

$$\text{Energy (Mev)} = \frac{\text{Range (grams/cm}^2) + 0.165}{0.0536}.$$

The gamma-ray, as is to be expected from a positron emitter, is absorbed to half-value by

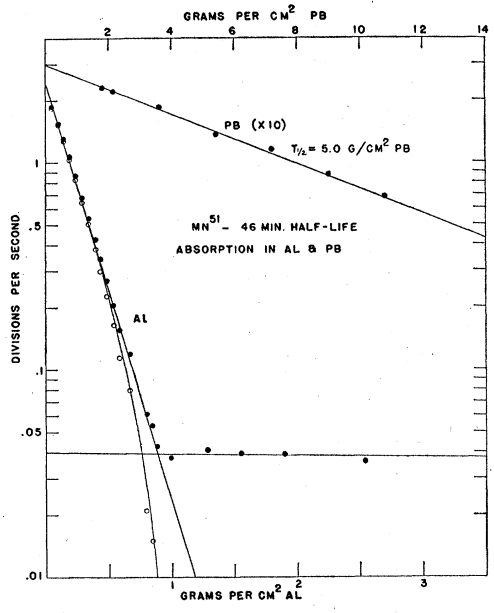

FIG. 3. Absorption curves of Mn^{51}.

5.0 grams/cm² Pb, which represents, according to the data of Gentner,[7] the 0.5 Mev annihilation radiation.

The aluminum absorption curve is somewhat anomalous, in that the softer component of the total radiation appears to be linear (on a semi-log plot). If the gamma-ray is subtracted, the curve for the particles is then concave towards the origin; this may indicate the presence of a homogeneous group of electrons. A possible explanation could be K electron capture as an alternative method of decay, followed by the emission of a gamma-ray that is highly internally converted.

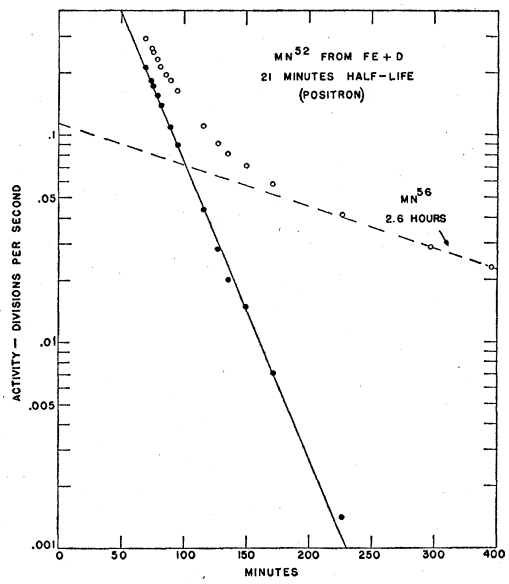

FIG. 4. Decay of Mn^{52}.

Mn^{52} FROM Fe+D: HALF-LIVES 21 ± 2 MIN. (+) AND 6.5 ± 1.0 DAYS (+)

A short account of manganese isotopes with half-lives 21 minutes and 6 days, formed by deuteron bombardment of iron, has already been given.[2] Both activities are positron emitters and chemical identification has been carried out. The 21-minute period has also been observed by Darling, Curtis and Cork[8] after the same type of bombardment.

Two positron emitting manganese isotopes, Mn^{52} and Mn^{54}, can be expected through the disintegration type $Fe(d,\alpha)Mn$; nevertheless, we believe both these activities must be described as isomers of Mn^{52}. This follows from the fact that neither period is obtained by deuteron bombardment of chromium, and the reaction $Cr(d,n)Mn$ could lead to Mn^{54} but not to Mn^{52}. Furthermore, neither activity has been found

[6] Widdowson and Champion, Proc. Phys. Soc. **50**, 192 (1938).
[7] Gentner, J. de phys. et rad. **6**, 274 (1935).
[8] Darling, Curtis and Cork, Phys. Rev. **51**, 1011 (1937).

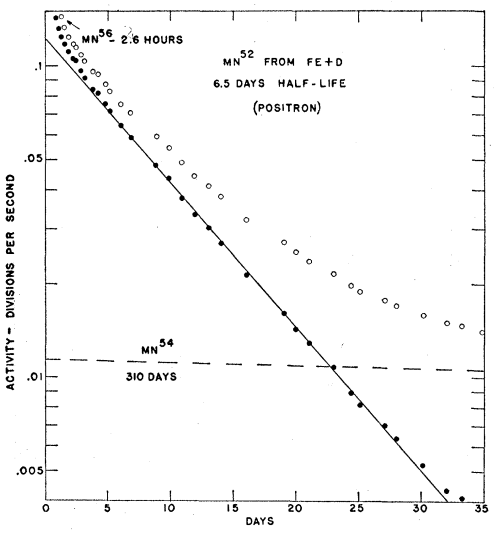

FIG. 5. Decay of Mn^{52}.

after the bombardment of manganese with the fast neutrons from Li+D, wherein the reaction $Mn^{55}(n,2n)Mn^{54}$ could be expected. And finally, this identification is consistent with our failure to detect either activity after neutrom bombardment of cobalt; if either were due to Mn^{54} the reaction $Co^{57}(n,\alpha)Mn^{54}$ might occur, in analogy with the known transmutation $Co^{59}(n,\alpha)Mn^{56}$. The scarcity of Co^{57}, however, could be used to vitiate this last argument.

We therefore conclude the two activities to be isomers, formed according to

$$_{26}Fe^{54}+{}_{1}D^{2}\rightarrow{}_{25}Mn^{52}+{}_{2}He^{4}$$
$$_{25}Mn^{52}\rightarrow{}_{24}Cr^{52}+{}_{+1}e^{0}\ (21\text{ minutes; }6.5\text{ days}).$$

The decay of the 21-minute positron activity is shown in Fig. 4. The manganese was separated from iron that had been exposed to 6 microamperes of 5.5 Mev deuterons for 10 minutes. Even with so short a bombardment, the 2.6-hour activity of Mn^{56} is relatively strong.

Figure 5 exhibits the decay of the 6-day positron emitter, intermixed with the 2.6-hour Mn^{56} and the 310-day Mn^{54} activities (see below). This sample was obtained from iron activated by a 4-hour bombardment with 15 microamperes of 5.5 Mev deuterons. The average value of the half-life, obtained from four specimens, is 6.5 ± 1.0 days.

The absorption in aluminum of this activity is given in Fig. 6. Inasmuch as the 310-day activity was also present during the lifetime of the 6-day period, this figure was obtained by correcting for the contribution of this long period to the composite absorption data obtained at the early age of a few days. The resultant curve is therefore subject to more than the usual amount of error, but suggests a range for the 6-day activity equal to 0.20 ± 0.05 gram per cm^2 Al, or a maximum energy for the positrons of about 0.7 Mev. The gamma-ray has not been examined but is probably the 0.5 Mev annihilation radiation.

Mn^{54} FROM Fe+D, Cr+D AND V+He: HALF-LIFE 310 ± 20 DAYS (K ELECTRON CAPTURE)

It has previously been shown by us[2] that a chemically identified manganese isotope of half-life several months is produced by deuteron bombardment of iron. Only electrons were reported to have been observed. More complete studies of this activity have been carried out on three samples that have been followed from 10 to 15 months, and we can now give the half-life more precisely as 310 ± 20 days. Absorption measurements indicate that the electrons are not disintegration particles, but are photoelectrons ejected from the extranuclear structure following "disintegration" of the radioactive nucleus by K electron capture, rather than by positron emission. This will be discussed more fully in a subsequent paragraph.

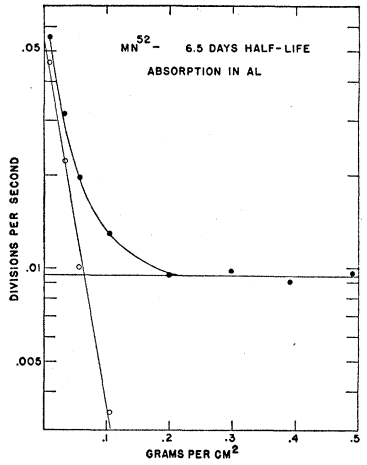

FIG. 6. Absorption curves of Mn^{52}.

Only two choices are available for this isotope when produced from iron by deuteron bombardment: Mn^{52} and Mn^{54}. We have given evidence above for the assignment to Mn^{52} of the isomeric activities with 21 minutes and 6 days half-lives, so that the 310-day period probably is to be associated with Mn^{54}. This supposition is confirmed by the fact that we have been able to produce the same activity, also chemically shown to be due to manganese, by activating chromium with deuterons. Here the reaction $Cr(d,n)Mn$ could lead to Mn^{54} but not to Mn^{52}. Additional evidence that this activity is identical with that produced from iron is the similarity of their absorption curves (see below).

The excessively long period is an adequate explanation for our inability to produce this activity through $Mn^{55}(n,2n)Mn^{54}$, and the scarcity of Co^{57} is an additional reason why the reaction $Co^{57}(n,\alpha)Mn^{54}$ has not been detected.

Study of the isotopic structure of the elements suggests that a conclusive identification of Mn^{54} should be obtained by alpha-particle bombardment of the single isotope of vanadium, since the only radioactive nucleus that could be formed by any known reaction should be Mn^{54}, through $V^{51}(\alpha,n)Mn^{54}$.

Ridenour and Henderson[4] have bombarded vanadium with 9 Mev alpha-particles and reported two activities with half-lives 1.2 minutes and about 67 minutes. No chemical separation was made. We have activated vanadium with 16 Mev alpha-particles and have obtained a decay curve showing a number of short periods and a very much longer activity, of much larger yield than the short periods, which is apparently going into the 310-day period to be expected on the basis of the data

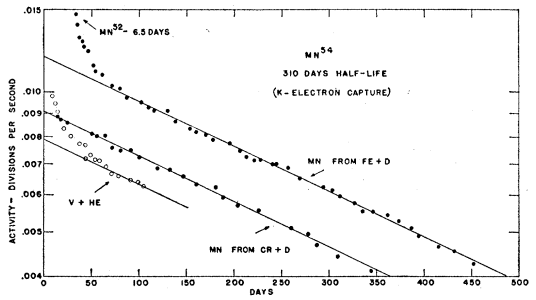

FIG. 7. Decay of Mn^{54}.

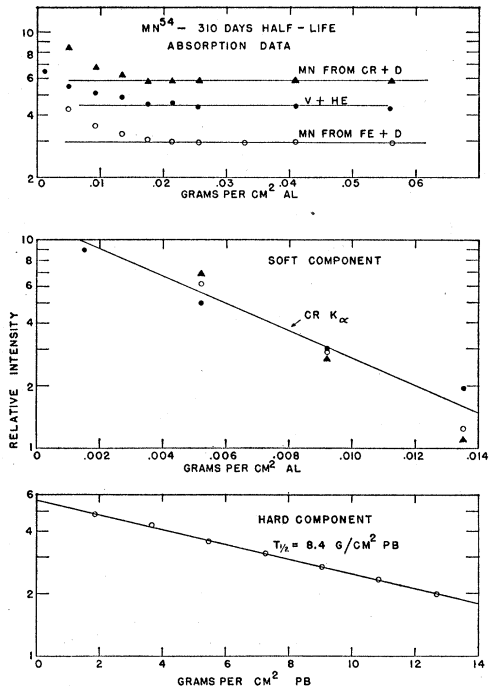

FIG. 8. Absorption curves of Mn^{54}.

given above. We have not been at liberty to perform a chemical analysis on the vanadium, which is supposed to be spectroscopically pure, so that this identification with Mn^{54} is somewhat speculative. Nevertheless, an absorption curve taken on the long-lived activity shows characteristics approximately identical with the 310-day manganese from the deuteron activated chromium and iron, so that we feel reasonably assured the assignment is justified. The other activities induced in the vanadium, might be due to impurities or possibly to neutrons (produced during transmutation of V^{51} to Mn^{54}) or to residual deuteron activation.

The 310-day period has therefore been produced by the transmutations

$$_{26}Fe^{56}+{}_{1}D^{2}\rightarrow{}_{25}Mn^{54}+{}_{2}He^{4},$$
$$_{24}Cr^{53}+{}_{1}D^{2}\rightarrow{}_{25}Mn^{54}+{}_{0}n^{1}$$

and possibly also by

$$_{23}V^{51}+{}_{2}He^{4}\rightarrow{}_{25}Mn^{54}+{}_{0}n^{1}.$$

Each is followed by

$$_{25}Mn^{54}+{}_{-1}e^{0}\rightarrow{}_{24}Cr^{54}\ (310\ \text{days}).$$

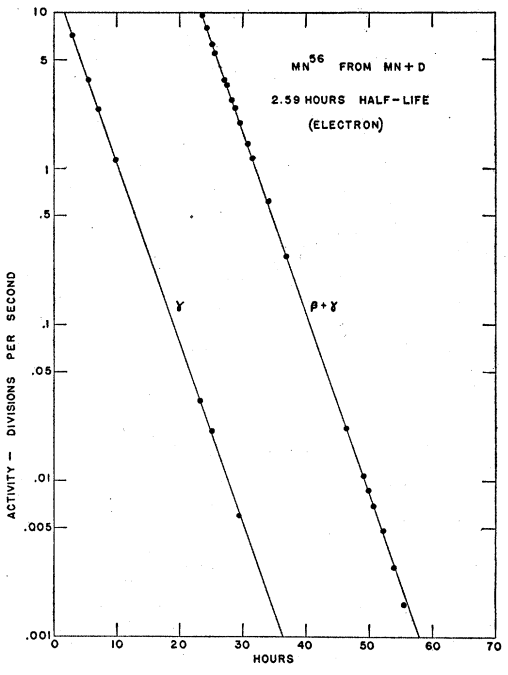

FIG. 9. Decay of Mn^{56}.

Figure 7 shows the decay of these long-lived samples. The iron was activated for 4 hours with 15 microamperes of 5.5 Mev deuterons, the chromium for 1 hour with 25 microamperes of 5.5 Mev deuterons and the vanadium for 2 hours with 0.1 microampere of 16 Mev helium ions.

The absorption data taken on the 310-day samples from Fe+D, Cr+D and V+He are decidedly anomalous, as may be seen in Fig. 8. Ordinarily the ionization due to the soft component (electrons or positrons) is 30 to 70 times as much as that due to the gamma-ray, when observed with an electroscope with a thin aluminum window (0.0001 inch). In the present instances, however, the gamma-ray causes more ionization than does the soft component. Absorption curves were taken on the manganese from an Fe+D bombardment with and without a magnetic field of 3600 gauss in the 10 cm gap between the active specimen and the electroscope; the identity of these curves indicated that there were no appreciable numbers of disintegration particles that were capable of penetrating the window. This suggests, on the basis of the work of Alvarez[9] with Ga^{67}, that the decay of Mn^{54} is neither by electron emission to Fe^{54} nor by positron emission to Cr^{54}, but rather that the Mn^{54} nucleus captures one of its extranuclear K electrons and thus becomes a nucleus of Cr^{54}. The vacancy in the K shell is then filled and the Cr $K\alpha$ x-ray line is emitted. The gamma-ray is only slightly internally converted.

The absorption curves of Fig. 8, although by no means a precise demonstration that this is the mechanism of decay, suggest that the soft components of all three specimens are indeed due to the $K\alpha$ line of chromium. The intensities of the radiations from the three samples have been adjusted to separate the curves in the upper part of the figure for greater clarity. In the central section, the intensities of the soft components (obtained by subtraction of their gamma-rays) have been adjusted to cluster about a line drawn with a slope of that of the Cr $K\alpha$ line.

The absorption in Pb of the 310-day activity from Fe+D is given in the lower part of Fig. 8. The gamma-ray is reduced to half-value by 8.4 grams/cm^2 Pb, which, according to Gentner's data,[7] indicates an energy of 0.85 Mev.

Mn^{56} FROM Mn+n, Mn+D, Cr+α, Fe+n, Fe+D, Co+n: HALF-LIFE 2.59±0.02 HOURS (−)

The identification with Mn^{56} of the much-studied 2.6-hour manganese period requires no argument at this date. It was originally produced by Fermi and his associates[10] through the reactions $Mn^{55}(n,\gamma)Mn^{56}$, $Fe^{56}(n,p)Mn^{56}$, and $Co^{59}(n,\alpha)Mn^{56}$. Recently Henderson and Ridenour[4] have obtained the same activity by $Cr^{53}(\alpha,p)Mn^{56}$. We have repeated all of these transmutations and in addition have produced the disintegrations $Mn^{55}(d,p)Mn^{56}$ and $Fe^{58}(d,\alpha)Mn^{56}$. Our best value for the half-life is 2.59±0.02 hours, obtained from a sample of manganese oxide that was bombarded for 1.5 hours with 8 microamperes of 7.6 Mev deuterons. This specimen was followed for 22 half-lives; the extrapolated initial intensity of the beta- and gamma-rays was 5×10^7 times background. The decay is shown in Fig. 9, wherein the gamma-ray curve was obtained by filtering through 0.2 cm Al and 1.1 cm Pb.

[9] Alvarez, Phys. Rev. **53**, 606 (1938).

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

A portion of the 2.6-hour activity observed after deuteron bombardment of iron must be ascribed to $Fe^{56}(n,p)Mn^{56}$, since the activity is found in iron sheets placed behind a thick iron target. The neutrons may come from the general background of neutrons always present during operation of the cyclotron, as well as from the $Fe(d,n)Co$ reaction.

Figure 10 exhibits the absorption curves in Al and Pb of Mn^{56}. Here again is the suggestion of a homogeneous group of electrons formed by internal conversion of a gamma-ray, since the curve for the electrons alone (obtained by subtracting the gamma-ray from the original data) is concave to the origin. The range is measured as 1.3 ± 0.1 grams per cm^2 Al, indicating a maximum energy of 2.7 ± 0.2 Mev. This is in fair agreement with the value obtained by more precise methods: cloud chamber measurements have given values of 2.8 Mev,[11] 3.2 Mev[12] and recently two groups of electrons have been reported[13, 14] at 1.2 and 2.9 Mev. These were detected by the same technique.

Our absorption data on the gamma-ray show it to be reduced to half-value by 11.4 grams per cm^2 Pb, which corresponds to an energy of 1.2 Mev by Gentner's compilation.[7] This is in only rough agreement with the value 1.65 Mev found by Mitchell and Langer,[15] who measured with coincidence counters the range of the Compton recoil electrons ejected by the gamma-ray.

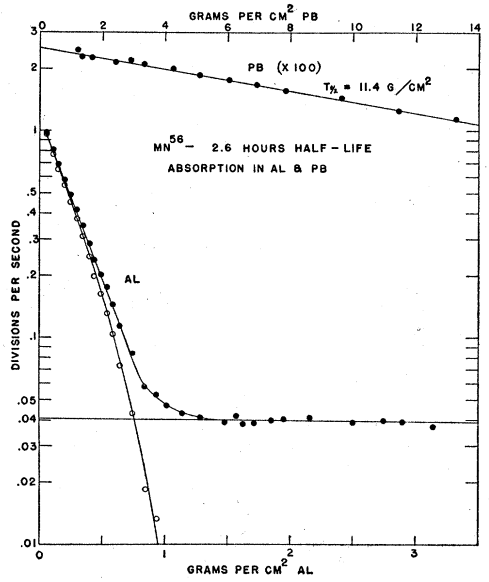

FIG. 10. Absorption curves of Mn^{56}.

TABLE I. *Transmutations and thick target yields.*

HALF-LIFE	REACTION	PROJECTILE ENERGY	PROJECTILES PER ACTIVE NUCLEUS
46 min.	$Cr^{50}(d,n)Mn^{51}$	5.5 Mev	2×10^6
21 min.	$Fe^{54}(d,\alpha)Mn^{52}$	5.5 Mev	1×10^8
6.5 days	$Fe^{54}(d,\alpha)Mn^{52}$	5.5 Mev	1×10^8
310 days	$Fe^{56}(d,\alpha)Mn^{54}$	5.5 Mev	2×10^8
310 days	$Cr^{53}(d,n)Mn^{54}$	5.5 Mev	1×10^6
310 days	$V^{51}(\alpha,n)Mn^{54}$	16 Mev	1×10^5
2.6 hours	$Mn^{55}(d,p)Mn^{56}$	5.5 Mev	1×10^6
2.6 hours	$Mn^{55}(d,p)Mn^{56}$	7.6 Mev	4×10^4

YIELDS

From the above data on bombardment time and current and initial activities, it is possible to make an estimate of the thick target yield for the different transmutations, with an approximate absolute calibration for the sensitivity of the electroscope. These computations (Table I) must be considered as only very rough, since no endeavor was made to obtain quantititative yield measurements. In the case of the 310-day activities, an estimate of the number of disintegrations was made from the ionization produced by the gamma-rays only (there were no particles present); for the other activities, the ionization due to both particles and gamma-rays was used. The yields are given as though the target element were composed entirely of the single isotope concerned.

This research has been made possible by the generous and continued support of The Research Corporation and The Chemical Foundation. The work was facilitated by W.P.A. assistance.

[11] Gaerttner, Turin and Crane, Phys. Rev. **49**, 793 (1936).
[12] Alichanian, Alichanow and Dzelepow, Nature **136**, 257 (1935).
[13] Brown and Mitchell, Phys. Rev. **50**, 593 (1936).
[14] Bacon, Grisewood and Van der Merwe, Phys. Rev. **52**, 668 (1937).
[15] Mitchell and Langer, Phys. Rev. **52**, 137 (1937).