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Proton Induced Radioactivity of Manganese*

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The reaction Cr^{52} (p, n) Mn^{52} at 6.6-Mev proton energy produces isomers with period 21.3 min. and 6.5 days. These are positron emitters with maximum positron energy of 2.2 Mev (21 min.) and 0.77 Mev (6.5 day). The gamma-ray spectrum for each isomer has been investigated. There are 2 gamma-rays per positron in the short period activity. The gamma-radiation in the 6.5-day activity is so intense that 95 percent of these disintegrations must be due to K-electron capture; this is followed by emission of probably two gamma-rays. In the remaining 5 percent the positron emission is followed by three gamma-rays. The threshold for the 21-min. activity and the yield for both periods have been determined.

SAMPLES of metallic chromium have been bombarded with 6.6-Mev protons produced in the University of Rochester cyclotron. Positron activities of two periods were observed and studied: 21.3 min. and 6.5 days. These periods have been observed in other reactions, and Livingood and Seaborg,[1] assign them to isomers of Mn^{52}. This conclusion is consistent with results to be presented here.

I. MEASUREMENT OF PERIODS

The bombarded samples of metallic chromium weighed about 5 g. The surface of the metal was cleaned by grinding on an emery wheel. The initial activity for the 21-min. period was 67 microcuries per microampere of protons for infinite duration of bombardment at 6.6-Mev proton energy. The initial activity for the 6.5-day period was 4.4 $\mu C/\mu a$ under the same conditions.

The two activities mentioned were identified chemically as Mn. A layer a few tenths of a millimeter thick was dissolved off of the Cr target by immersion in hot HCl. To the acid solution $MnCl_2$ was added for a carrier. NaOH was added until the solution was basic, and it was then oxidized with Na_2O_2. The solution was boiled, diluted, and filtered. The precipitate showed the 21-min. and 6.5-day activities.

The periods were measured with an ionization chamber containing Freon-12 at 2 atmospheres pressure and an FP-54 amplifier arranged for automatic photographic recording of the data.[2] Figure 1 shows the decay curve of a Cr target which had been exposed to 6.8-Mev protons at 0.07 μa for an hour. From this curve we determine the shorter period as 21.3 min. Subtracting the long period activity from the total, we see that the two periods mentioned account for practically all of the observed activity, and there is no

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* A preliminary report of this work was made at the Washington meeting of the American Physical Society, December 1938. See Phys. Rev. **55**, 604 (1939).

† A report of work done at the University of Rochester during the summers of 1938 and 1939.

[1] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 391 (1938).

[2] S. W. Barnes, Rev. Sci. Inst. **10**, 1 (1939).

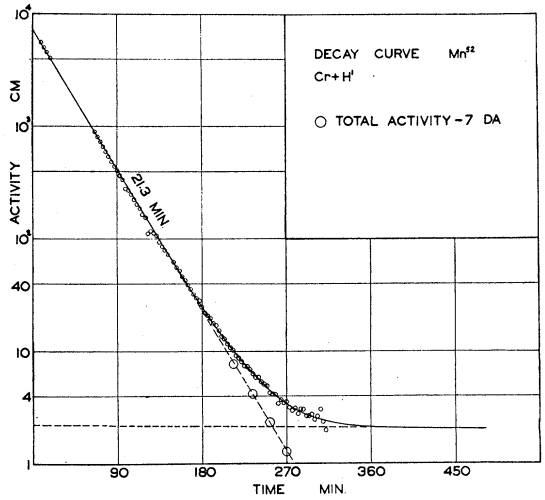

FIG. 1. Decay curve of Mn^{52}. Cr target bombarded at 6.8 Mev for 60 min. at 0.07μa. Measurements start 4 min. after end of bombardment.

evidence for a 46-min. period, which activity occurs at lower proton energies.[3,4] We have also found the 310-day period assigned by Livingood and Seaborg[1] to Mn^{54}, but have not studied it. Figure 2, showing the decay curve of a Mn precipitate several days after bombardment of the parent Cr, gives the half-life as 7.4±1 days. The Cr target was bombarded for 45 min. at a current of 0.5 μa of 6.6 Mev protons. This period is slightly greater than Livingood and Seaborg's value of 6.5±1 days, and since the 310-day activity was not subtracted, it seems reasonable to assume 6.5 days to be the more precise value.

II. Positron Energy

The energies of both sets of positrons were measured by means of a cloud chamber in a magnetic field. Histograms are shown in Fig. 3 and Fig. 4. The cloud-chamber tracks were mostly positrons, distributed in the usual beta-ray spectrum. The upper energy limits of the positron spectra are 2.2 Mev (21 min.) and 0.77 Mev (6.5 day). The energy 0.77 Mev agrees fairly well with the value (0.7) given by Livingood and Seaborg.[1] They do not report the energy of the 21-min. positrons. We have measured the range in Al of these particles and find it to be 0.973 g/cm^2. Using the relation[5]

$$\text{energy} = (\text{range (g/cm}^2) + 0.165)/0.536 \text{ Mev},$$

we get a value of 2.12 Mev.

III. Excitation Function

By bombarding a pile of thin films of chromium the threshold for the 21-min. activity was determined. The intensity of the 6.5-day activity was far too small for such a measurement. Cr films about 0.18μ thick were evaporated onto 2.5μ Al foil. The Cr side of the foil was exposed to the proton beam so that the Mn recoil nuclei could not get away. The stopping power of the pile of eight foils was measured by placing it in the proton beam and observing the decrease in range of the protons. The stopping power of the pile was 6 cm air equivalent. Protons of this energy produce no activity in Al of such a period and intensity to be troublesome even in comparison with the activities of these thin targets. The incident proton energy at the first foil was 6.6 Mev. There was an 0.05 Mev energy

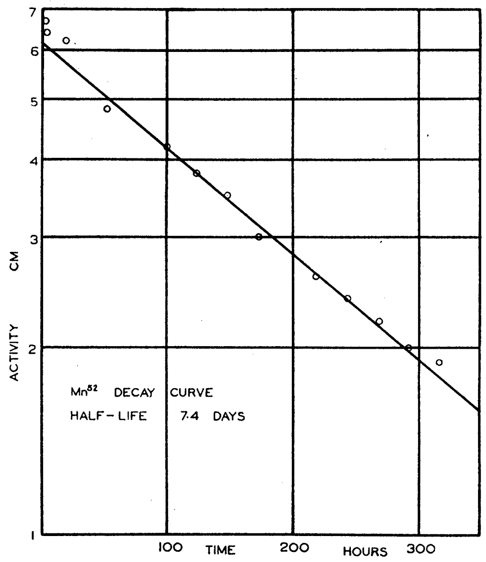

FIG. 2. Decay curve of Mn precipitate. Cr target bombarded at 6.6 Mev for 45 min. at 0.5μa. Measurements start two days after end of bombardment.

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[3] Barnes, DuBridge, Wiig, Buck and Strain, Phys. Rev. **51**, 775 (1937); DuBridge, Barnes, Buck and Strain, Phys. Rev. **53**, 447 (1938).

[4] Delsasso, Ridenour, Sherr and White, Phys. Rev. **55**, 113 (1939).

[5] E. E. Widdowson and F. C. Champion, Proc. Phys. Soc. **50**, 192 (1938).

loss in each Cr+Al film. The decay curves for the various films are shown in Fig. 5. The initial activities are plotted against the proton energy in Fig. 6. We find the threshold for the 21-min. activity to be 6.3 Mev. The actual reaction energy (threshold corrected for nuclear recoil) is $6.3(52/53)=6.2$ Mev. All we can say about the threshold of the 6.5-day activity is that it does not differ from 6.2 Mev by more than a few tenths of a Mev; at energies lower than 6.3 Mev the 46-min. and 310-day activities are so great compared to the 6.5-day that a more precise determination of this threshold is hardly feasible.

IV. Gamma-Rays

Gamma-ray absorption curves are shown in Fig. 7. The first part of the curve for the 21-min. period has a slope which probably corresponds to annihilation radiation. In the 6.5-day activity the intensity of annihilation radiation is negligible compared to that of higher energy radiation. Using data given by Gentner[6] we estimate the average gamma-ray energies as 1.1±0.2 Mev (21 min.) and 0.9±0.2 Mev (6.5 day). Since both gamma-ray spectra are probably complex these energy values are very uncertain.

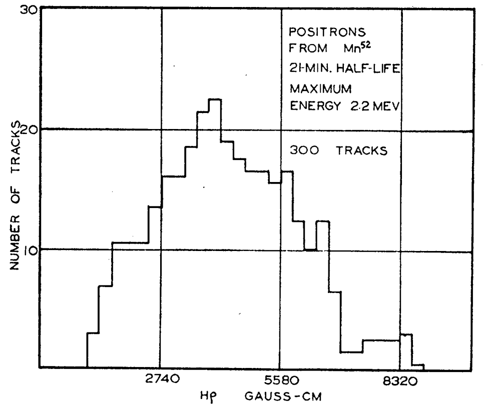

FIG. 3. Distribution of positron tracks from 21-min. Mn^{52}.

The source used in plotting the gamma-ray absorption curves (Fig. 7) was a thick Cr target covered with an Al sheet. Thus positrons emitted in any direction were stopped, and for each positron there must be two quanta of annihilation radiation. The ionization current due to the hard component is twice that due to the soft (from Fig. 7), and the ionization per quantum of the hard radiation will possibly be greater than that of the 0.5-Mev radiation. This suggests that there may be about two 1.1-Mev quanta per positron. The assumption that there are two 1.1-Mev gammas per positron is also necessary to account for the observed threshold of 6.2 Mev, since the upper limit of the positron spectrum is only 2.2 Mev, leaving 2.2 Mev for the gamma-ray energy.

We can estimate the intensity of the gamma-radiation in the 6.5-day activity by calculating the ratio $\Gamma=\gamma/(\gamma+\beta)$ of ionization produced by gammas to total ionization, the former being measured with an Al absorber thick enough to stop all positrons.

These ratios are 0.025 (21 min.) and 0.24 (6.5 day). For the 21-min. activity $\gamma/\beta=2.5$ percent, of which two-thirds is for hard gamma-rays. For these only, $\gamma/\beta=1.6$ percent. For the 6.5-day activity $\gamma/\beta=32$ percent. Since the sensitivity of the ionization chamber does not vary greatly with energy in this region, and since the gamma-rays have nearly the same absorption coefficients, the number of gammas per positron in the 6.5-day activity is $32/1.6=20$ times as many in the 21-min. activity, which means 40 gammas per positron.

We conclude that the 6.5-day isomer decays not only by positron emission, but also by *K*-electron capture followed by emission of gamma-radiation. Presumably 3 gammas accom-

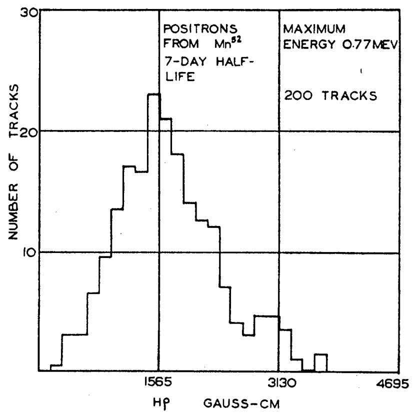

FIG. 4. Distribution of positron tracks from 6.5-day Mn^{52}.

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

pany the positron, leaving the other 37 for K capture. Assuming 2 gammas per K capture, then there are about 18 K captures per positron, or positron emission occurs in only 5 percent of the disintegrations.

We have looked for x-rays emitted by the 6.5-day isomer. There is evidently a soft radiation, which we were unable to identify, apparently because of the absorption in air.

V. Coincidence Counter Measurements

The data presented so far show that there might be some interesting features of the disintegration of the 6.5-day isomer, and it seemed at least possible that these could be investigated by means of coincidence counters. Thick-walled alcohol-argon filled counters were used, and an amplifier with a resolving time (τ) of $(9.17\pm0.25)\times10^{-9}$ min. The details of the counters, amplifiers, scaling, circuit, and the technique of measuring τ will be described elsewhere. Counter data were taken for both short and long periods. In each case, measurements were made of single and coincidence counting rates with the thin (0.5 mm) source in both positions A and B of Fig. 8. In position A the source is placed between two Al slabs 0.25 in. thick which serve to stop all positrons. In position B ideally only half of the positrons produce annihilation radiation which affects the counters. Actually, the number of annihilation quanta counted in position B is more than half of the total number, and this ratio must be ascertained by separate measurements.

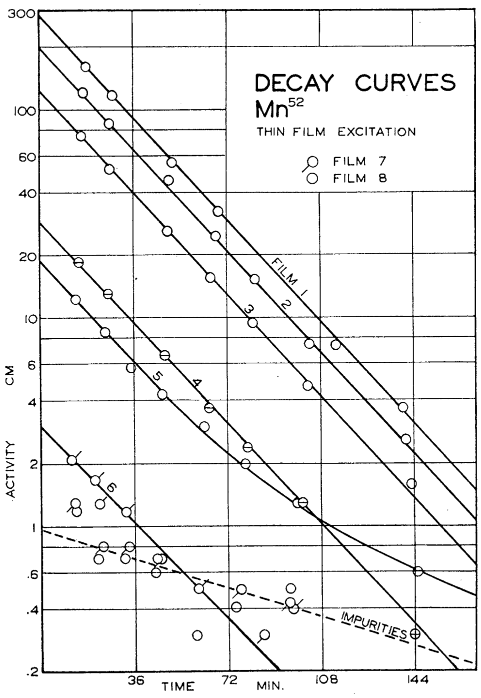

Fig. 5. Decay curves of thin films of Cr bombarded with protons.

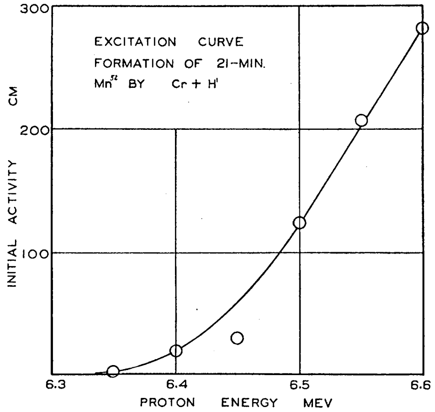

Fig. 6. Excitation curve for the formation of the 21-min. Mn^{52}. Threshold estimated at 6.3 Mev.

If we let N_1 = single counting rate, position A; N_2 = single counting rate, position B; N_3 = coincidence counting rate, position A; N_4 = coincidence counting rate, position B; N = number of disintegrations per minute; n = average number of gamma-rays per positron (exclusive of annihilation radiation); S_γ = average gamma-ray sensitivity of the counter including the solid angle; S_a = annihilation radiation sensitivity of counter; and K = a constant to be determined later (with the source in a vacuum, $K=1$), we can write the following equations for the counting rates:

$$N_1 = NnS_\gamma + 2NS_a, \tag{1a}$$

$$N_2 = NnS_\gamma + KNS_a, \tag{1b}$$

$$N_3 = NS_\gamma^2 n(n-1) + 4NnS_\gamma S_a, \tag{1c}$$

$$N_4 = NS_\gamma^2 n(n-1) + 2KNnS_\gamma S_a. \tag{1d}$$

To determine K, separate measurements were made using a source of Zn^{63}. This element emits 2.3-Mev positrons (nearly the same positron energy as the 21-min. Mn^{52}) and no gamma-rays,

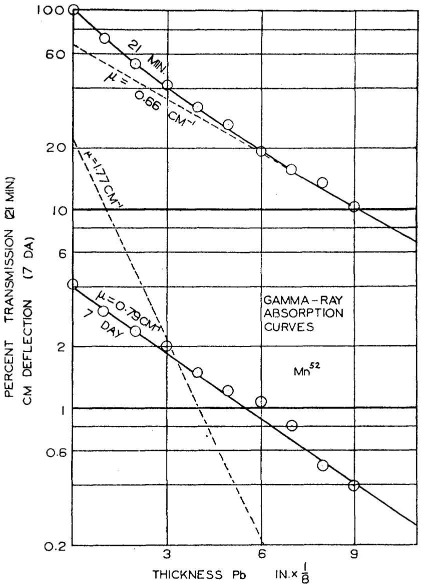

FIG. 7. Absorption curves in Pb of gamma-radiation from Mn^{52}, both isomers.

so that $n=0$. By measuring single counting rates in positions A and B we can solve Eqs. (1a) and (1b), obtaining $K=2N_2/N_1$. The result of such measurement is $K=1.06$.

We can now solve Eqs. (1) for n, N, S_a, S_γ. We are at present interested in the value of n:

$$n=\left[1-2\frac{2N_4-KN_3}{2N_2-KN_1}\cdot\frac{N_1-N_2}{N_3-N_4}\right]^{-1}.$$

In measuring N_1 and N_2 the background of the counters must be subtracted as well as the activity of the 6.5-day Mn^{52} present. The single counting rates of the two counters were nearly the same, and for N_1 and N_2 averages of the rates for the two counters were used. Actually, N_1 and N_2 could not be measured until the coincidence counting rates were too small to measure. In measuring N_3 and N_4 the procedure was to start with a sample which gave about 1000 coincidence counts per min. in position A. Coincidence decay curves in positions A and B were then plotted (with cosmic-ray background subtracted). Since N_1, N_2, and τ are known, the accidental counting rates are known, so one can calculate the true initial coincidence counting rates. The first part of the data (counting rate 1000 coincidences per min.) is useless, for these are mostly accidental coincidences. At best, the method does not yield data of high precision with a source which has a 21-min. period, for the interval of time in which coincidence counts can be made is too short. The data for 21-min. Mn^{52} are as follows:

$N_1=395500\pm120$ (counts/min.)
$N_2=305500\pm90$
$N_3=360\pm17$ (counts/min.)
$N_4=231\pm11$.

The fractional probable errors in N_1 and N_2 were computed in the usual way: 0.67 divided by the square root of the total number of counts. Since N_3 and N_4 are differences of measured and accidental coincidence rates, the probable error was computed for each measured point, and the values of N_3 and N_4 are means weighted according to the inverse square of the probable error. Using Eq. (2) we find $n=1.26\pm0.17$. This suggests that positron emission is followed sometimes by the emission of two 1.1-Mev gamma-rays, and at other times by one 2.2-Mev gamma. Presumably the high energy gamma-radiation is not very intense, for it did not show up at all in the absorption curve (Fig. 7). There is some further evidence for the higher energy gamma-rays in cloud-chamber pictures which show pair production by this radiation in a 2.5-mil Pb foil.

The 6.5-day isomer, which disintegrates 95 percent of the time by K capture, can be investigated by comparison of the ratios N_3/N_1 and N_4/N_2 for the two isomers. Taking averages of the data for each isomer, we have

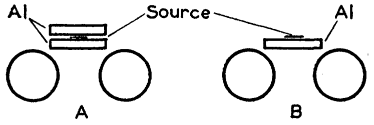

FIG. 8. Geometry of counters and source.

ISOMER	N_3/N_1	N_4/N_2
21-min.	$(10\pm0.5)\times10^{-4}$	$(7.5\pm0.3)\times10^{-4}$
6.5-day	$(3.5\pm0.08)\times10^{-4}$	$(2.9\pm0.1)\times10^{-4}$.

Thus the coincidence rate for this activity is less than half that for the short period activity. Remembering that in the 21-min. activity there are 3.3 gammas per disintegration (including annihilation radiation) and never more than 4, it appears that for the majority of 6.5-day disintegrations there are at most 2 gamma-rays. For disintegrations in which there is positron emission the energy available for gamma-radiation is 3.6 Mev. If some of this is the same as the 2.2-Mev radiation of the 21-min. isomer, the energy of the additional gamma-ray is 1.4 Mev. The lower absorption curve of Fig. 7 gives the energy of the 6.5-day gamma-radiation, as about 1 Mev. If there were only one gamma-ray of this energy per *K* capture, an implausibly large amount of energy would have to be assigned to the neutrino. It is more probable that there are two gammas per *K* capture, one of them being possibly of about 3.2-Mev energy.

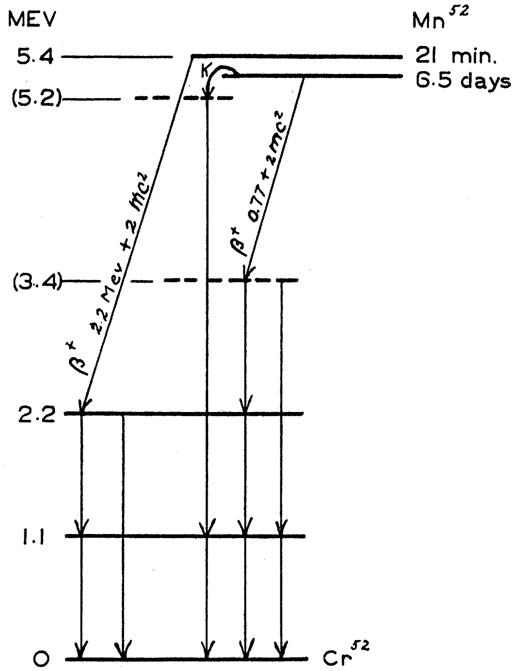

FIG. 9. Energy level diagram consistent with the information available on the isomers Mn^{52}. Uncertain values of energy are in parentheses.

VI. YIELD

The initial activity of the 6-day isomer of 4.4 μC/μa is given in Section I. The initial activity due to positrons only is $0.75\times4.4 = 3.3$ μC/μa. If all the disintegrations were by positron emission the initial activity would be $3.3/0.05 = 66$ μC/μa. Using the calibration data of the ionization chamber, we get the yields for each isomer at 6.6-Mev proton energy:

21 min.	0.40×10^{-6}	radioactive atoms per proton
6.5 day	0.68×10^{-6}	radioactive atoms per proton.

VII.

The data on the two isomers of Mn^{52} are summarized in the following table:

ISOMER	DISINTEGRATION	β^+ *KE*	THRESHOLD	YIELD
21.3-min.	β^+	2.2 Mev	6.2 Mev	0.40×10^{-6}
6.5-day	β^+ 5% *K* 95%	0.77		0.68×10^{-6}

The results of the gamma-ray measurements are somewhat uncertain, but all available data are consistent with the following possible scheme: the 21-min. isomer disintegrates with the emission of a positron followed by two 1.1-Mev gammas or one 2.2-Mev gamma. The 6.5-day isomer, disintegrating by positron emission emits 3.6 Mev of energy, probably in 3 quanta; disintegrating by *K* capture, it emits 4.2 Mev of radiation in two quanta, one of which must be about 1 Mev. An energy level diagram corresponding to this scheme is shown in Fig. 9.

In conclusion I wish to thank Professor L. A. DuBridge for making available laboratory facilities at the University of Rochester, and I am indebted to the whole staff at Rochester for their generous assistance, in particular Dr. S. N. Van Voorhis, Dr. S. W. Barnes and Dr. C. V. Strain. I wish also to thank Dr. L. M. Langer of Indiana University for many helpful discussions regarding the counter work.