Half-Life of Mn⁵³

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A value of 3.7×10⁶ yr ($\pm 10\%$) for the half-life of Mn⁵³ was determined by a direct method involving measurement of both the specific activity [(dis/min Mn⁵³)/(μ g Mn)] and the isotope ratio $(Mn⁵³/Mn⁵⁵)$. The Mn⁵³ specific activities were determined by neutron activation, and the Mn^{55}/Mn^{53} isotopic ratios by mass spectrometry. A surface-ionization method was applied using a rhenium single filament with the sample mounted in a silica-gel layer. This technique gave a stable and enhanced ion beam of Mn^{+} . One source of Mn^{53} was an electroplated Cr target bombarded by 15-MeV α particles in a cyclotron. The other source of Mn⁵³ was the Mn fractions extracted from iron meteorites by carrier-free processes. Activities of 10 dis/min $\rm Mn^{53}$ and 2000 dis/min $\rm Mn^{54}$ were obtained with about 2 $\rm \mu g$ of $\rm Mn^{55}$ from the Cr target. The isotopic ratio Mn⁵⁵/Mn⁵³ was 750. Activities of about 1 (dis/min Mn⁵³)/(μ g Mn) and isotopic ratios Mn^{55}/Mn^{53} of 3000-4000 were obtained from the iron-meteorite extracts.

I. INTRODUCTION

The half-life of the long-lived nuclide Mn^{53} has recently been studied by several methods. The published values for the half-life, however, are still scattered in a wide range between $10⁶$ to $10⁷$ yr with high uncertainties. A reliable figure for the half-life of Mn^{53} will provide one of the most important items of basic data in the studies of current cosmochemical problems involving the fossil records of natural nuclear interactions found in extraterrestrial materials over time scales of the order of the long-lived nuclides Be¹⁰ ($T_{1/2}$ = 2.7 × 10⁶) yr), Al^{26} (7.4×10⁵ yr), and other nuclides.¹

In general, the most reliable result for this class of long-lived activities is obtained by a direct method that includes an absolute disintegration measurement by radiation counting, and an isotopic-ratio measurement of the radioactive isotope relative to some stable isotope by mass spectrometry. Some serious difficulties, however, arise with this method for Mn^{53} . The preparation of suitable samples to be tested, the determination of a very low content of Mn⁵³ relative to Mn⁵⁵, and a very low specific activity of Mn⁵³ relative to Mn^{54} ($T_{1/2}$ = 303 days), an interfering by-product in artificial samples, all offer serious problems.

The indirect methods have given some indications of the half-life. From the considerations of the nuclear spin and nuclear-reaction yield data, a value for the half-life was estimated by Sheline and Hooper² to be 2×10^8 yr. Although this value seems to be the correct order of magnitude, it has been used widely for a long time for lack of other strong empirical estimates.

Some stone meteorites which have been irradiated by galactic cosmic rays for not more than

 $10⁷$ yr contain Mn⁵³ activity as high as other meteorites of much longer exposure. This indicates that secular equilibrium with the decay rate or activity equal to the production rate has already practically been obtained and hence that the halflife of Mn^{53} is short compared with 10^7 yr. On the other hand some iron meteorites which have sufficiently long terrestrial ages so that much of the Cl^{36} has decayed, still contain high Mn⁵³ activities. This indicates that the half-life of $Mn⁵³$ is long compared with 3×10^5 yr, probably longer than 10^6 yr. From such cosmochemical evidence, Kaye and Cressy³ estimated $(1.9 \pm 0.5) \times 10^6$ yr as the best estimate for the half-life of Mn^{53} , based on radiochemical data of particular meteorites. The reliability naturally depends upon the radiochemical determinations of Mn⁵³, Be¹⁰, and other isotopes in the meteorites, and these determinations may not always be accurate enough to extend this argument.

Matsuda, Umemoto, and Honda⁴ recently reported on another indirect method. Since the activities of Mn^{53} and Mn^{54} are almost nearly equal at the time of fall for meteorites of long cosmicray ages, the production ratio of Mn^{53}/Mn^{54} from high-energy spallation on Fe is nearly unity. Assuming that the production ratio is the same for artificial bombardment by 730-MeV protons, the activity ratio of Mn^{53}/Mn^{54} in the target will be the inverse ratio of the half-lives of Mn^{53} and Mn^{54} . Using a neutron-activation technique^{5, 6} to determine the Mn⁵³ and Mn⁵⁴ activities in a 9-yr-old iron target which had been bombarded by 730- MeV protons, Matsuda, Umemoto, and Honda' calculated a value for the half-life of Mn⁵³ of $(2.9 \pm 1.2) \times 10^6$ yr. It should be noticed that these two estimates are both based upon the assumption

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of constant cosmic-ray flux.

For the direct empirical method, the Mn extracts from iron meteorites provide useful samples. The Mn extracts from the Canyon Diablo iron meteorites were examined by Hohlfelder' using mass spectroscopy. The mass spectrometer has a high-abundance sensitivity employing a double-focusing system. This result, $(10.7 \pm 4.5) \times 10^6$ yr, was the longest half-life yet given for this nuclide. The high value is hardly understood from the above-mentioned cosmochemical considerations. If the difficulty is experimental, it could be mentioned that the sample employed was of a very low specific activity in Mn^{53} ; ca. 0.06 (dis/ min Mn⁵³)/(μ g Mn).

In view of the earlier studies and their discrepancies, it became clear that a sample of a higher specific activity should be prepared for mass spectrometry and, at the same time, the Mn^{54} in the sample must be low enough to allow a neutronactivation measurement of Mn⁵³.

As was suggested in our previous paper, 6 we extracted such Mn samples from two sources. One was an artificial, α -ray-bombarded chromium target. The target by-product, Mn^{54} , was fairly low, only about 10^2 times the Mn⁵³ activity. The presence of such a level of Mn⁵⁴ causes no difficulty in the Mn^{53} activity measurement. By using the Mn⁵³ (n, γ) reaction, about 10³ times the activation of Mn^{53} into Mn^{54} is routinely accessible using a reactor with about 3×10^{18} n/cm² total dose. Contamination of Mn in the chemical extractions was limited to the order of 10^{-6} g. The final extracts were about 4 times superior

to any of the meteorite extracts, the other source of Mn⁵³, which is described below.

From iron meteorites about 0.3 to 0.4 (dis/min of $Mn^{53}/(g$ meteorite) were extracted along with 0.2 to 0.3 ppm Mn^{55} as was reported in the previous paper.⁶ This contamination of Mn has been found to be unavoidable and fairly constant among meteorites. The level of spallation products which have accumulated in space during the exposure age of several hundred million years must be at least ~ 0.1 ppm. About 1 (dis/min Mn⁵³)/ $(\mu g$ Mn) are therefore available almost independent of the meteorite sample.

The limitation to the specific activity is obvious for the meteorite samples, but a much higher specific activity is available from artificial bombardment. Both sources seem to be useful for the determinations, and the Mn^{55}/Mn^{53} ratios have been estimated in the range from 0.7×10^3 to 4×10^3 . Ratios in that range can be determined by ordinary settings of a conventional single-focusing mass spectrometer with a reasonable accuracy when any impurity such as $Cr⁵³$ does not seriously interfere with the mass spectra.

II. EXPERIMENTAL

A. Preparation of the High-Specific-Activity Samples

1. The artificial Mn^{53} sample. As was already reported in our previous paper⁶ a chromium target was bombarded by 15.2-MeV α particles. Mn⁵³ was produced by the Cr⁵⁰(α , *n*) and Cr⁵⁰(α , *p*) reactions. The target was a thick copper plate with a thin, 70-mg/cm2, electroplate of natural

TABLE I. Determination of Mn⁵³ activity by neutron activation in α -bombarded Cr target. Sample: aliquots of Mn extract from 15.2-MeV α -bombarded Cr target (No. 2), a total about 2 μ g Mn was found in the extract; irradiation: JRR 3 VG, 273.5 h (Japan Atomic Energy Research Institute, Tokai-mura, Japan); total flux: $1.3 \times 10^{18} n/cm^2$. All $Mn⁵⁴$ activities were corrected for decay to the date of neutron irradiation. Errors indicate 1 standard deviation. Mn⁵⁴ γ -ray counting was performed with a $1\frac{3}{4}-in$ -diam $\times 1\frac{3}{4}-in$. (1.3-cm-diam $\times 2.0$ -cm well) NaI(Tl) detector, counting efficiency (835-keV photo peak): 8.8%, background: \sim 1 counts/min.

² 216 \pm 9 on the date of α bombardment. Total 11.0 dis/min Mn⁵³ and 2370 dis/min Mn⁵⁴ were found to be produced after the α bombardment.

 b Mn⁵⁴ added as tracer.</sup>

chromium, 2 cm by 6 cm. The α beam hit an elliptical area of 0.5 cm by 3 cm. The bombardment was carried out using a cyclotron at the Institute of Physical and Chemical Research, Saitama, Japan. The beam current was about 20 μ A and the total dose was 1.9 C or 6×10^{18} α particles during the continuous 25-h run. From this run, we expected a production of about 10 dis/min Mn⁵³, assuming $T_{1/2} = -4 \times 10^6$ yr, 400 mb for the effective mean cross section and 22 mg/cm' Cr for the available thickness of the target. The production of Mn⁵⁴ as a by-product, by $\overline{C}r^{52}(\alpha, d)$ Mn⁵ reactions where $Q = -10.6$ MeV, was extremel low, with an effective cross section far lower than 1 mb in this energy region.

After bombardment the target was dissolved in HCl and a carrier-free extraction of Mn was performed. A two-stage anion-exchange separation of Mn from Cr was performed in concentrated HCl solution. The activity of Mn'4 was useful as a tracer in the extraction steps.

According to neutron-activation analysis 11 dis/ min of Mn⁵³ were produced and most of it was recovered in the extracts along with 2000 dis/min of Mn^{54} and about 2 μ g of Mn (Table I).

Because the presence of $Cr⁵³$ interferes directly with the determination of Mn^{53} in the mass spectrometry, a thorough removal of chromium from the Mn fraction was necessary. For this purpose the last steps of the purification were carried out as follows. The pH of the solution was adjusted to about 9 by an acetate and tartrate mixture after addition of ascorbic acid. The alcoholic solution of TTA was added and the Mn-TTA complex was extracted into ethylacetate. The complex was back-extracted with dilute HNO, . The nitric acid solution was evaporated and the ammonium salts were decomposed with *aqua regia*. The Mn fraction was dissolved in a few drops of $5 M HCl + 40\%$ isopropyl alcohol solution and was adsorbed on a 2-ml column of anion-exchange resin. The column was washed with 4 ml of the $5 M HCl + 40\%$ isopropyl alcohol solution and Mn was eluted with 2 ml of 6MHCl.

Finally the Mn fraction was evaporated in a small (1- to 2-ml capacity) silica crucible and 60% HC104 was added dropwise. After each drop of $HClO₄$ was added the crucible was heated to eliminate $HClO₄$ and to decompose the Mn salt to the form of a stain of brown oxide. In this final step, which was repeated three times, the last trace of Cr could be removed from the sample. Using a $Cr⁵¹$ tracer, the effectiveness of the removal was monitored. After the removal of the trace of chromium by HClO, treatment the residue in the silica crucible was converted to a soluble salt form by a gentle heating with HC10, . Flame spectrophotometry was applied for the Mn analysis in the extract. The over-all recovery of purified Mn from the target was about 55%.

2. The natural Mn^{53} sample. Several iron-meteorite specimens were used as a source for the natural Mn⁵³. The activity of Mn⁵³ has already been determined in several meteorites, and thus suitable specimens could be chosen. The meteorites Thunda, Treysa, Grant, and Carbo were used.⁸ A carrier-free recovery was performed similar to that performed on the artificial target sample described above. A carrier-free Mn⁵⁴ tracer (200-500 dis/min) was used to determine the chemical yield. The 5-10-g meteorite specimens were dissolved in aqua regia. Isopropyl ether was used to extract iron, and anion exchange in HCl solution was performed to separate Mn from Co, Ni, and other metals. Further purification by cation exchange with elution in 1 M HCl was followed by anion exchange in the isopropyl alcohol system, etc., described previously. The chemical yields ranged from 70 to 90% . The content of Mn⁵³ in the final extracts was directly determined by neutron activation. Atomic absorption was also applied for the Mn analysis. The specific activties of the extracts were estimated both from chemical-recovery data and from direct-activation analyses.

B. Mass Spectrometry of the Mn Samples

An AEI-MS 5 surface-ionization-source mass spectrometer, $R = 30$ cm and 90° deviation, was used with an electron multiplier attached to the collector, a digital voltmeter with a sensitivity of $1 \mu V$ (6 digits), and a printer. Usually the output of the voltmeter was printed every 0.2 sec to give peak-height readings. To obtain a stable and enhanced ion beam of Mn', the sample was supported with silica gel placed on a rhenium ribbon $(0.75 \cdot m \times 0.25 \cdot mm)$ filament.^{9, 10} By this method with $mm \times 0.25$ -mm) filament.^{9, 10} By this method with 0.1 μ g of Mn a sufficiently stable ion beam of 10^{-14} to 10^{-13} A was available for at least a fe 10^{-14} to 10^{-13} A was available for at least a few hours. This technique was also found to be useful in eliminating chromium interference with increasing filament current. The peak heights of Cr^{52} decreased to less than 10^{-3} of the Mn⁵⁵ peak. Any organic compound peak at $M/e = 53$ seems to be reduced to an insignificant level in the presence of $SiO₂$ gel. A high resolution of about 600 was obtained with a narrow collector slit of 0.² mm. Without the silica-gel support, using a triplefilament setting composed of two Ta filaments for the side and a Re for the center, any hydrocarbon contributions appearing at 53, such as the C_4 group compound, was separated from the top of the $Mn^{53} + Cr^{53}$ peak at 52.95 by a distance of about 1.⁵ times the width of half maximum of the metal peak.

One serious problem in this experiment was the appearance of a faint unidentified background or ghost at $M/e = 53$. In the presence of more than 0.1 μ g Mn, this background, however, was less than 10% of the signal of Mn⁵³ appearing in meteorite sample. The background contribution did not seem constant throughout the run, which started with a lower filament current such as 2.0 A (1000'C as measured by an optical pyrometer) and ended with 2.5 A (1150°C) or a higher current. In this work, this background contribution caused the largest uncertainty in the isotopic measurements. The contribution usually decreased gradually with time. The origin was certainly neither from any organic materials composed of H, C, N, and/or O, nor from ClO⁺, Cd⁺⁺, Pd⁺⁺, or ZrO⁺⁺.

The amounts of Mn in the meteorite samples mounted were sometimes as low as $0.1 \mu g$. In such cases a small reduction for the 53 peak should be made. It was simply assumed that the background contributions were variable from 2 \times 10⁴ to 2 \times 10⁵ in the 55/53 ratios with \pm 100% errors corresponding to from 0.1 to 1 μ g Mn mounted. With smaller size samples with 55/53 ratios much higher than $10⁴$, this ambiguity might have been very serious.

Some examples of the mass-spectroscopic data are tabulated in Table II, and an example of the

mass spectra is shown in Fig. 1. The data indicate that the peak-height ratios I^{55}/I^{53} observed were essentially independent of measuring conditions or stages of the run. The $Cr⁵³$ contribution to the $Mn^{53} + Cr^{53}$ peak was calculated from the observed Cr^{52} peak height assuming a fixed ratio for the Cr.

III. RESULTS

The results obtained in this work are all sum-The results obtained in this work are all sum-
marized in Table $III.^{11}$. In this table, any systemat ic absolute error in the Mn⁵³ activity standardization and the systematic mass-discrimination effect were not taken into account. From the chromium target sample, a value of 3.6×10^6 yr was obtained with a high reproducibility. From meteorite Mn samples, figures of about 5% higher were obtained. The errors attached to the meteorite sample values were also somewhat higher than for the chromium target values. The essential agreement, however, found in two entirely different sources, artificial and natural, seems to be excellent.

In addition to the statistical error shown in the tables, there are two possible sources of systematic error in our value of the Mn⁵³ half-life. The first is associated with the determination of the specific activity of Mn⁵³ in dis/min μ g. The calculation of the specific activity required the

Filament current (A)	No. of peak group ^a	I^{55}/I^{52} (Mn^{55}/Cr^{52})	Peak-height ratios I^{55}/I^{53} b $(Cr^{53}$ subtracted)	Representative value for I^{55}/I^{53}
Cr target, $0.4 \mu g$ Mn				
No. 2				
2.15	15	$60 - 80$	730 ± 60	750 ± 30
2,25	10 ₁₀	~100	746 ± 40	
2,40	15	~150	757 ± 32	
2,50	9	$200 - 350$	743 ± 30	
2,60	8	~1000	730 ± 46	
2.75	$\overline{\mathbf{4}}$	~1000	$~1770$ (unstable)	
Thunda, 1μ g Mn				
(iron meteorite)				
2,20	15	$350 - 600$	3700 ± 300	3700 ± 200
2.30	21	$200 - 1700$	3600 ± 250	
2.42	10 ₁₀	$1600 - 2000$	3770 ± 200	
Reagent Mn, $0.5 \mu g$ Mn				
(chemically processed) ^c				
2.05	$\overline{\mathbf{4}}$	$~1$ $~200$	$\geq 10^4$	
2,12	8	$200 - 500$	$2 \times 10^4 - 10^5$	
2.25	3	$500 - 1000$	$2 \times 10^5 - 3 \times 10^5$	$\geq 10^5$
2,35	6	$5000 - 8000$	$10^5 - 2 \times 10^5$	
2,48	7	$~10^{4}$	$10^5 - 10^6$	

TABLE II. Examples of mass spectrometry of Mn samples.

^a One peak group includes one full return, up and down, magnetic mass scan between 52 and 55.

^b Errors indicate 1 standard deviation.

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Passed through cation-exchange and anion-exchange separation steps.

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measurement of a Mn⁵³ reference sample of known activity and an analysis of the content of Mn in the various sample solutions.

Our Mn⁵³ reference was extracted from the Grant iron meteorite¹² and was calibrated by direct x-ray counting using a Mn^{54} source supplied by the Laboratoire Métrologie de la Radioactivité, France. The source was guaranteed to be accurate to $\pm 1.5\%$. Our calibration error was estimated as 5%. For the chemical analysis of Mn, we assumed an error of 3% which includes all possible systematic errors.

The second source of systematic error is associated with the mass-spectrometric measurements. An error of 2% is related to the massdiscrimination effect of the instrument, conventionally expressed as $(55/53)^{1/2}$. In addition, we have assigned a conservative error of 2% to the peak-height-ratio determinations. We use this error simply because of uncertainties in measuring the narrow peaks of the higher abundance ratio. Both errors must be increased because of an additional factor arising from subtraction of the Cr^{53} contribution to the $M/e = 53$ peak. Usually the Cr⁵³ contribution was of the same order or less than that of Mn⁵³. This means that the estimated error for mass discrimination and peak height should be about 4% each.

Based on the above considerations, we estimate $±8\%$ as the systematic error in this experiment. Combining this systematic error with the statistical error of 6@ shown in Table III, we may express the half-life of Mn⁵³ as

 $T_{1/2}(\text{Mn}^{53}) = (3.7 \pm 0.4) \times 10^6 \text{ yr}.$

IV. DISCUSSION

In this work a value of 3.7×10^6 yr was obtained for the half-life of Mn^{53} by direct measurements. This figure does not agree with any of the previous estimates, except the one of $2.9 \pm 1.2 \times 10^6$ yr which

FIG. 1. Example of mass spectra of $Mn⁵³$. Sample: Cr target No. 2; amount of Mn mounted: ca. $0.4 \mu g$; specific activity of the extract: ca. 6 (dis/min Mn⁵³)/ $(\mu g$ Mn); filament: rhenium ribbon, 2.7 A; electron-multiplier gain: 4×10^4 ; peak at $M/e = 53$: ca. 90% Mn⁵³ + ca. 10% $Cr⁵³$; Mn⁵⁵/Mn⁵³ observed: 750 ± 30. For actual peak top readings a slower scan speed was employed, and discontinuous scannings were performed.

was estimated by the authors based on an indirect method.⁴ A new figure obtained in this work may be applied directly in several current investigations in the fields of nuclear- and cosmo-chemistry. The following may be mentioned as directly relating to the topics in the latter field

A. Fossil Records of Nuclear Interactions in Extraterrestrial Materials

 $Mn⁵³$ will inform us of a history up to about 5 $\times10^6$ yr, the mean life, in any extraterrestrial sample. The history is then the oldest among the records of the long-lived nuclides such as Be¹⁰, Al^{26} , and others. In this respect, the comparisons of Mn⁵³ content with the solar cosmic-ray track observations found in the surface of luna
rocks may be interesting.¹³⁻¹⁵ rocks may be interesting.

B. Mass Spectrometry of Mn⁵³

Routine Mn⁵³ measurements in iron meteorites will be possible with 1-g samples. For this purpose, however, a more extensive purification of silica gel for mass spectrometry might be useful. This method is more sensitive than direct lowbackground x-ray counting, though less sensitive than the neutron-activation method.

On the other hand, for the samples which contain a high Mn^{54} activity, such as artificial samples extracted from bombarded targets where the activity ratio of Mn^{53}/Mn^{54} may be in the vicinity of 10^{-6} , the determination of the Mn⁵³ is best done by mass spectrometry when the Mn^{55} contamination is small.

C. Cross-Section Determinations

For the reaction $Mn^{53}(n, \gamma)Mn^{54}$, a figure of 350 $± 50$ has already been obtained for the "activation factor,"⁶ a practical term, expressed in $\frac{dis}{min}$ factor,"⁶ a practical term, expressed in (dis/mir
Mn⁵⁴)/(dis/min Mn⁵³) per $10^{18}~n_t/\rm cm^2.^{16}$ From the $T_{1/2}$ of Mn⁵³, $\sigma(n, \gamma)$ is directly calculated to be 80 ± 10 b. When we take 360 ± 30 for the "activation factor" from the more accurate data obtained in our current experiment, we obtain $\sigma = 82 \pm 7$ b.

our current experiment, we obtain $0 - 62$
In our previous report,⁴ relative spallatio yields for Mn^{53} and Mn^{54} were measured in an iron target bombarded with 730-MeV protons. Using the new $T_{1/2}$ for Mn⁵³, we obtain $\sigma(Mn^{53}) = 37 \pm 15$ the new $T_{1/2}$ for Mn⁵³, we obtain $\sigma(Mn^{53}) = 37 \pm 15$
mb in contrast to $\sigma(Mn^{54}) = 30$ mb.¹⁷ Though a relatively high uncertainty exists, spallation cross sections were found to be essentially the same for Mn^{53} and Mn^{54} , as assumed in our previous report.

For the calculation of Mn^{53} production by spallation and by lower-energy reactions with protons, direct cross-section measurements with accelerators will be informative. For this the half-life is used directly in providing the absolute crosssection data.

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 $0.693\times$ (No. of Mn⁵³ atoms disintegration rate of Mn⁵³ $1.445{\times}10^{10}~{\rm yr}$ (specific activity) \times (Mn⁵⁵) /(Mn⁵³) $^{\circ}$ where the specific activity is in units of dis/min of Mn^{53} per μ g Mn⁵⁵, and (Mn⁵⁵)/(Mn⁵³) is the relative atomic abundance.

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