Half-lives of ^{44}Ti and ^{207}Bi

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(Received 26 January 1990)

An end-window gas-flow proportional counter system, including an automatic precision sample changer, was used to measure the decay of radiations from two samples of ^{44}Ti and one of ^{207}Bi. Comparison counts were taken on a ^{36}Cl standard, and the ratio of counts determined the decay. Points consisted of alternate 30-min counts on each sample and on the standard, totaling 30 h on each, and were taken at intervals of about one month. Based on 3 years of counting on the ^{207}Bi source and on one of the ^{44}Ti sources, and 1 year on the second ^{44}Ti sample, half-lives were obtained as follows: ^{44}Ti, 66.6(16) yr and ^{207}Bi, 34.9(4) yr. The ^{207}Bi value lies between the two most recently reported values. In the case of ^{44}Ti there are discrepancies with all three previous results including an Argonne accelerator mass spectroscopy value of 54.2(21) yr.

I. INTRODUCTION

^{44}Ti is one of 18 radioactivities found in meteorites,[1] presumably having been produced by the cosmic-ray spallation of iron. As one of their uses,[1] these activities can give information on variations in cosmic-ray flux. Another use[1] of ^{44}Ti is to deduce the terrestial age of a meteorite on a time scale of of a few hundred years from the ^{44}Ti radioactivity level, in a manner analogous to ^{14}C dating. There are two applications in the field of γ-ray astronomy, in one of which ^{44}Ti is currently a prime subject of searches[2] in the remnants of supernova 1987A as well as in older supernovae. Already the 847- and 1238-keV γ-ray lines from 77-day ^{56}Co have been observed[3] from supernova 1987A. After the ^{56}Co decays sufficiently it is hoped that the 1157-keV γ-ray line from ^{44}Ti-^{44}Sc will be identifiable. If this becomes possible it could help in a more detailed understanding of the supernova process. All of the existing ^{44}Ca is believed to have originated from ^{44}Ti decay following nucleosynthesis. In the other application, ^{56}Co and ^{44}Ti are thought[4] to be the only two important sources of positrons from explosive nucleosynthesis, as observed from a gamma-ray peak at 476 keV thought to be associated with positronium.

All such studies depend on having a reliable and accurate value for the half-life of ^{44}Ti. Three measurements have been reported thus far, all of them absolute determinations of the decay constant. Two of these were made in 1965 using dilution counting techniques and a mass spectrometer to obtain isotope ratios. Wing *et al.*[5] obtained 46.4(17) yr and Moreland and Heymann[6] found 48.2(9) yr. More recently, the Argonne National Laboratory group of Frekers *et al.*[7] used the accelerator mass spectrometer (AMS) technique to obtain isotopic ratios in their sample. By combining that result with a counting measurement they obtained a half-life of 54.2(21) yr for ^{44}Ti.

In view of our earlier work[8] at Brookhaven on the half-life of ^{32}Si [$T_{1/2}=172(4)$ yr], measured by its direct decay over a 4-year period, it was suggested to us by Kutschera that a similar measurement could be done on ^{44}Ti. The Argonne group, in fact, very kindly furnished us with a portion of their activity.

Another radioisotope in which we have long had an interest is ^{207}Bi, which was discovered in 1951 by Neumann and Perlman[9] and estimated by them to have a half-life of approximately 50 yr. A much shorter half-life, 8.0(6) yr, was measured by Cheng *et al.*[10] Subsequent results[11–16] were in the range 28–38 yr, the most recent being those of Yanokura *et al.*[15]—33.4(8) yr, and Hoppes *et al.*[16]—32.2(13) yr. The Yanokura *et al.* value was revised[17] to 36.7(14) yr because of a corrected ^{211}Po α-particle branching ratio. (The uncertainty of 1.4 yr was calculated by the present authors by combining the various reported uncertainties.) Because of the wide range and inconsistencies of these various results, we decided to make a study of ^{207}Bi concurrently with ^{44}Ti. The half-life of ^{207}Bi is not believed to be of particular significance, but this activity is a convenient γ-ray energy and intensity standard whose half-life ought to be established with greater accuracy and reliability.

II. EXPERIMENTAL METHODS AND RESULTS

An end-window gas-flow proportional counter system, including an automatic precision sample changer, was used for our measurements. This equipment has been thoroughly described,[18] and its use in measurements on ^{32}Si has been given in detail.[8] Samples for the present work were secured to standard brass holders with Duco cement. They were then covered with 0.25-mg/cm^2 thick Al foil, attached with an electrically conducting cement, in order to eliminate erratic counting effects due to source charging. In the previous experiments[8] on ^{32}Si small but statistically significant periodic annual variations in the ratios of counts were observed and were thought to be caused by humidity and temperature variations in the air path between the sources and the detector, even though the box pressure was accurately controlled. The earlier micrometer settings for the distances

between the sources and the end-window counter were 1.0 mm for the ^{32}Si source and 4.0 mm for the stronger ^{36}Cl source. For the present experiments the test samples were all the 1.0-mm setting and a similar but weaker ^{36}Cl source was used that allowed it to be brought to a micrometer setting of 1.0 mm giving the desired counting rate. Thus the direct air path for the ^{36}Cl comparison source was decreased by 3 mm and this was expected to reduce the periodic variation effect. In fact, no periodic annual variations in the ratios of counts seemed to be evident in the present results covering approximately 3 years of measurements.

The present procedures were nearly the same as in the ^{32}Si runs, i.e., after a 1-day equipment warmup, the first ^{44}Ti sample was counted for 20 cycles of 30-min per cycle, alternated with 30-min counts on the ^{36}Cl reference source ($T_{1/2}=301{,}000$ yr), altogether totaling 10 h on each. Three such runs were averaged and the ratio of counts gave the plotted points. After 3 days of such runs, it was exchanged for the ^{207}Bi source, for 3 days, and then to the second ^{44}Ti source for 3 days beginning in the third year of counting. Individual runs were rejected if the box-pressure regulator lost control due to very high or very low barometric conditions, and make-up runs were made.

A. ^{44}Ti

The first ^{44}Ti source consisted of activity provided by the Argonne National Laboratory group. The sample had been produced prior to 1980 via the ^{45}Sc($p,2n$)^{44}Ti reaction at $E_p=45$ MeV in the Julich cyclotron. After 3 years of cooling, chemical separation and purification was performed.[7] The upper part of Fig. 1 shows the 32 ^{44}Ti/^{36}Cl data points from this sample recorded over a total of 3 years beginning in 1986. In all cases of Figs. 1–4 the ordinate is linear for convenience in plotting. Two changes of counting level occurred during the three years. As may be seen in the singles counts of Fig. 2, slight decreases in both counting rates took place at around $T=1.7$ yr and this resulted in breaks in the continuities of both the ^{44}Ti and ^{36}Cl curves. From our previous extensive studies[8] of the effects of changes of various parameters (amplifier gain, bias level on output pulses, counter voltage, counter gas-flow rate, and box pressure), we are confident that these small changes would have a negligible effect on the ratio of ^{44}Ti/^{36}Cl counts. In fact, no deviations were noticeable at $T=1.7$ yr in either the ^{44}Ti/^{36}Cl ratio in the upper part of Fig. 1 or in the ^{207}Bi/^{36}Cl data discussed below.

The second and much larger abrupt change in counting level occurred at around $T=2.85$ yr and this amounted to a decrease of about 0.5% in singles counting rates. An attempt was made to compensate for this by adjusting the bias on the output pulses so as to bring the ^{36}Cl singles rate back to very nearly its initial value. It took nearly 6 more months of counts before it was fully realized that the ^{44}Ti/^{36}Cl ratio points, beyond the time $T=2.85$ yr indicated by the arrow in Fig. 1, no longer matched the first 2.3 yr of data but fell somewhat lower than expected. The procedure was therefore to analyze the two regions

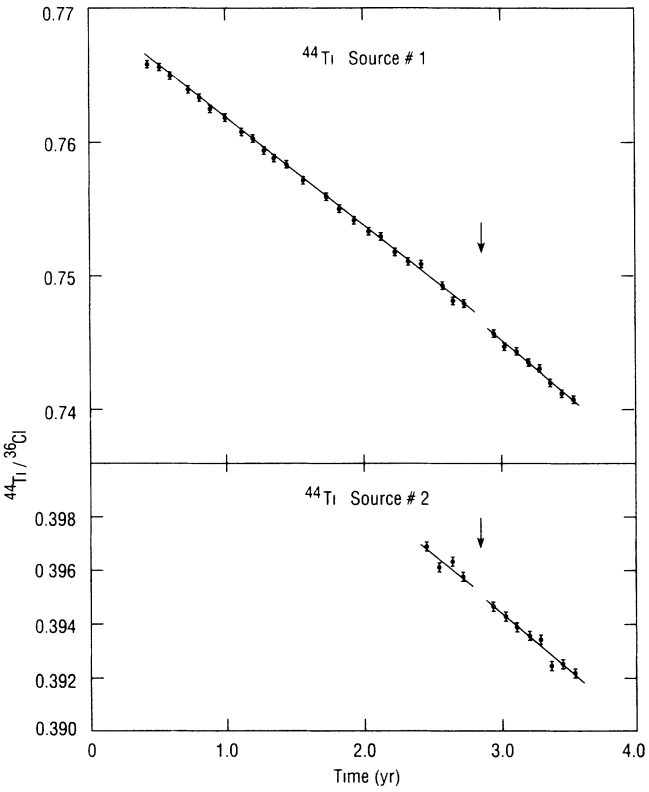

FIG. 1. Upper part, ratio of ^{44}Ti/^{36}Cl counts from the first ^{44}Ti sample measured for 32 points over a period of 3.1 yr. Points are averages of 3 runs, each with 10 h on each sample. Error bars are statistical. A change in counting level, indicated by the arrow at $T=2.85$ yr, made it necessary to analyze the data in two parts, as described in the text; lower part, ratio ^{44}Ti/^{36}Cl counts from the second source. Lines show the fits to the various regions.

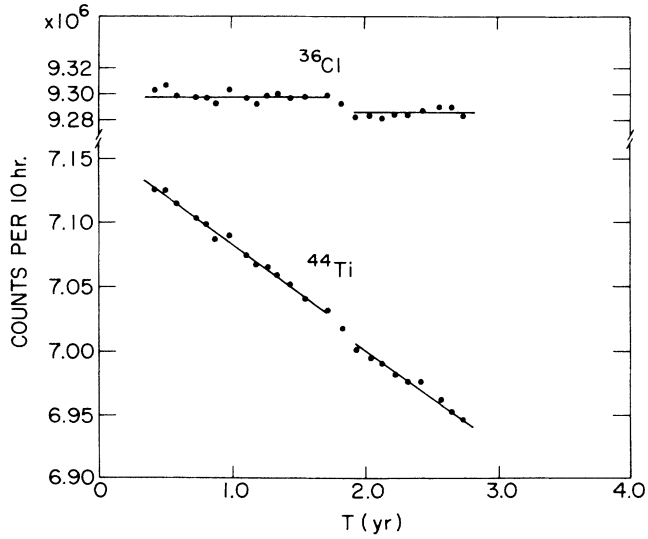

FIG. 2. Lower part, ^{44}Ti singles counting rate from the first sample measured over a period of 2.3 yr. Upper part, corresponding ^{36}Cl singles counts. Small shifts in both counting levels are apparent at about $T=1.7$ yr. Lines are hand-drawn guides to the eye.

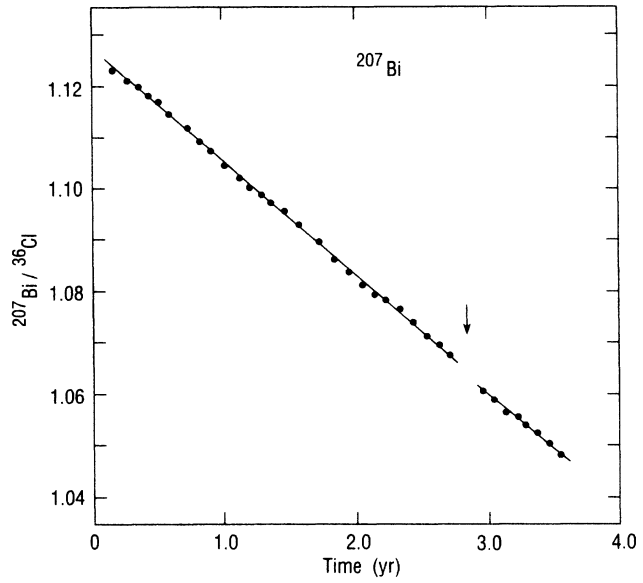

FIG. 3 Ratio of ^{207}Bi/^{36}Cl counts measured for 35 points over a period of 3.4 yr. Points were recorded as for ^{44}Ti in Fig. 1. Due to the counting level change, indicated by the arrow at $T=2.85$ yr, the data were analyzed in two parts. The lines show these two fits.

separately with the results shown in Table I. These fits were individually quite satisfactory since the uncertainty and standard deviation were 0.54 and 0.55 yr, respectively, for the first 24 points, and 2.80 and 2.89 yr, respectively, for the next 8 points. On the other hand, the single fit to all 32 points was poor in that the uncertainty was twice as great as the standard deviation. Because of a

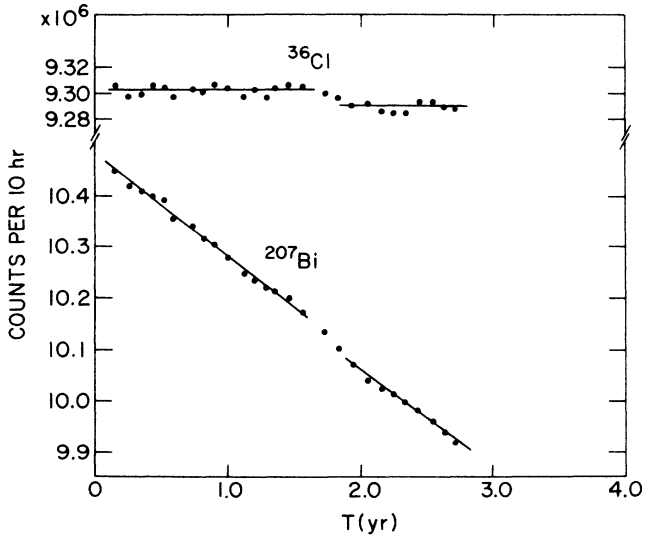

FIG. 4. Lower part, ^{207}Bi singles counting rate measured over a period of 2.6 yr. Upper part, corresponding ^{36}Cl singles counts. Small shifts in both counting levels are apparent at about $T=1.7$ yr. Lines are hand-drawn guides to the eye.

TABLE I. ^{44}Ti half-life results from analyses of the data in Fig. 1. Uncertainties are standard deviations.

^{44}Ti sample	Region of fit	$T_{1/2}$ (yr)
No. 1	First 24 points	66.9(6)
	Next 8 points	60.8(29)
No. 2	First 4 points	78.0(16)
	Next 8 points	62.0(36)
	Weighted average	66.6(8)

failure of the timer control unit and the need for extensive repairs, it was decided to terminate the experiments at $T=3.6$ yr.

When it became suggestive after 2 years of counting on the first ^{44}Ti source that the half-life was going to be longer than the value 54.2(21) yr reported by Frekers *et al.*,[7] we considered possible causes for the difference. The most obvious reason for this would be the presence of a long-lived contaminant. Assuming that 54 yr were to be the more nearly correct value, it is easy to show that in order to obtain a fitted value of ≈ 67 yr over a 3-yr counting period the contaminant contribution would have to be about 18% of the total activity. We therefore measured the γ-ray spectrum from the Argonne sample by means of a Ge(Li) detector system. The net spectrum after subtraction of room background showed only the x rays following electron capture, and the γ rays of 68, 78, 511, 1157, 1499, and 2656 keV due to the ^{44}Ti-^{44}Sc-^{44}Ca decay chain. Over the energy range up to 3 MeV no other lines were observed and an upper limit of $<1\%$ was placed on the relative intensity of any contaminant γ-emitting radioactivity.

The β-ray spectrum from the Argonne ^{44}Ti sample was then examined by means of a plastic scintillator. This showed the expected β with an end-point energy of 1476 keV plus a lower energy Compton-electron component due to the response of the detector to the 1157-keV γ rays. There was no evidence for a long-lived β-emitting contaminant. However, the selectivity here is not very good and we could only say that any long-lived β-emitting contaminant would have to be $<20\%$ of the total activity. From the process[7] by which the ^{44}Ti source was made there is no known pure β emitter that could be present and interfere with our measurements.

In spite of our conviction that the ^{44}Ti results from the first source were valid, we were sufficiently concerned about the discrepancy with the AMS value[7] that we initiated a second set of measurements on ^{44}Ti. An old source of ^{44}Ti was found among samples that had been accumulated by Sunyar. Although the exact origin of this source is not known, its γ-ray spectrum indicated a high degree of ^{44}Ti purity.

Data on the ^{44}Ti/^{36}Cl ratio using the second ^{44}Ti source are shown in the lower part of Fig. 1. Only 4 points were taken before the change of counting level occurred, indicated by the arrow at $T=2.85$ yr. Table I gives the results of fits to the separate regions of ^{44}Ti/^{36}Cl points before and after the level change occurred. The weighted average of the two results from the second ^{44}Ti source alone is $T_{1/2}=63(4)$ yr.

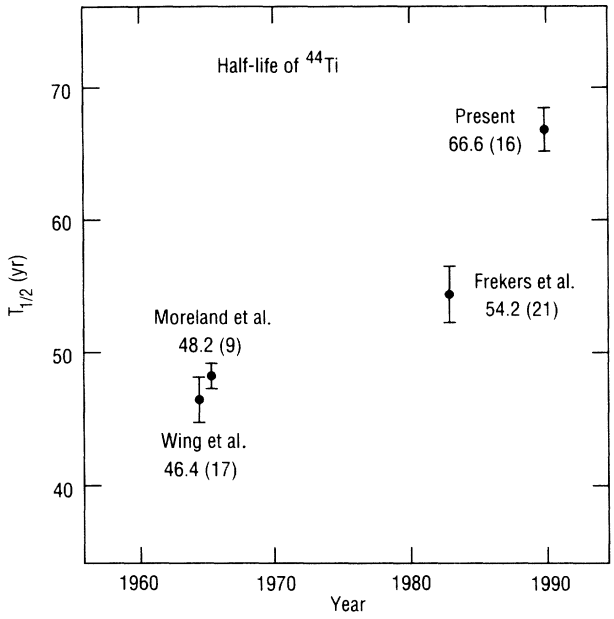

FIG. 5. Reported half-lives of ^{44}Ti vs the year of publication.

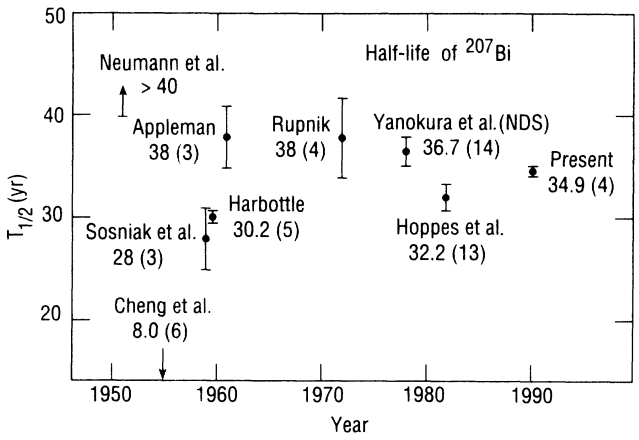

FIG. 6. Reported half-lives of ^{207}Bi vs the year of publication.

B. ^{207}Bi

Our source of ^{207}Bi dates from 1953 when a substantial supply of this activity was produced by bombarding a target of ordinary Pb with 22-MeV deuterons from the Oak Ridge National Laboratory cyclotron. Following a chemical separation at Brookhaven numerous electroplated sources were made.[19] Figure 3 shows the 35 ^{207}Bi/^{36}Cl data points recorded over a period of 3.4 yr. As in the ^{44}Ti case, the breaks in the singles rates of the ^{207}Bi and ^{36}Cl points in Fig. 4 are evident at around $T=1.7$ yr, although no deviation is apparent in the ratio data of Fig. 3. Again, because of the second and much larger shift at $T=2.85$ yr, indicated by the arrow in Fig. 3, the ratio data were analyzed in two separate parts. Results for the ^{207}Bi half-life were 34.88(21) yr from the first 27 points and 35.2(9) yr from the next 8 points. A single fit to all 35 points was poor, as in the ^{44}Ti analysis.

III. DISCUSSION

The separate results from the two ^{44}Ti sources, as given in Table I, are in agreement. Although the uncertainty for source #2 is relatively large, it is consistent with there being no long-lived contaminant in source #1. We adopt $T_{1/2}=66.6(16)$ yr for ^{44}Ti, where the uncertainty is taken as 2σ in order to cover possible systematic effects. In Fig. 5 the various values reported for the ^{44}Ti half-life are plotted versus the year of publication. We offer no explanation for the large inconsistencies of the various values. Thus our value differs from the Argonne result[7] by 6 times the uncertainty of their measurement. Assuming that the present result is valid, there would be some concern about the reliability of the AMS technique in this instance. Evidently more measurements would be desirable because of the importance of the activity as mentioned earlier. In the decay scheme of ^{44}Ti, as most recently studied,[20] the log ft value for the main $0^+\rightarrow 0^-$ 99.3% electron capture branch changes from 6.537(6) to 6.534(11) as a result of the presently adopted value for the half-life.

Our adopted value for the ^{207}Bi half-life is 34.9(4) yr where the uncertainty is again taken as 2σ. This is shown together with previous results, lying in the range 28–38 yr, in Fig. 6. Our result lies between the corrected value of 36.7(14) yr due to Yanokura *et al.*[17] and the value 32.2(13) yr due to Hoppes *et al.*;[16] it differs from both by amounts equal to or somewhat more than the sum of the uncertainties.

ACKNOWLEDGMENTS

We wish to thank E. K. Warburton for assistance with computer analysis. Research has been performed under Contract No. DE-AC02-76CH00016 with the United States Department of Energy.

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