Half-life of ^{44}Ti

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We present the results of two separate measurements of the half-life of ^{44}Ti relative to those of ^{22}Na and ^{207}Bi, respectively. By comparing the numbers of 1157-keV γ rays from ^{44}Ti to those of 1274-keV γ rays from ^{22}Na observed from a mixed source over a period of approximately 2 yr, we determined the half-life of ^{44}Ti to be 61.5 ± 1.0 yr. From an approximately 1-yr-long study of another mixed source, where we compared the numbers of 1157-keV γ rays from ^{44}Ti to those of 1064-keV γ rays from ^{207}Bi, we determined the ^{44}Ti half-life to be 62 ± 5 yr. From these two results, we have obtained a best value of 62 ± 2 yr for the half-life of ^{44}Ti. The astrophysical implications of this result are discussed. [S0556-2813(98)00704-3]

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I. INTRODUCTION

The long-lived radioisotope ^{44}Ti is of considerable interest in astrophysics. The relevant portion of its decay scheme and that of its daughter ^{44}Sc are shown in Fig. 1 [1]. ^{44}Ti decays to ^{44}Sc, emitting γ rays of 68 and 78 keV. ^{44}Sc subsequently decays with a 3.9-h half-life to ^{44}Ca, emitting an 1157-keV γ ray. ^{44}Ti is one of the few long-lived γ-ray-emitting nuclides expected to be produced in substantial amounts during a supernova explosion [2,3]. Its characteristic 1157-keV γ ray was observed from the young supernova remnant Cassiopeia A (Cas A) [4]. More recent studies of Cas A have provided indications of the 68- and 78-keV γ rays from ^{44}Ti as well [5]. ^{44}Ti can also be produced in meteorites through cosmic-ray interactions, thus providing information on solar activity from the cosmic-ray exposure of such objects [6]. Furthermore, the solar system abundance of ^{44}Ca is believed to have originated from the nucleosynthesis of ^{44}Ti and the subsequent decays [7].

In order to deduce the mass of ^{44}Ti ejected in the explosion using the γ-ray flux measured from a supernova remnant, one needs to know its age and distance as well as the half-life of ^{44}Ti. For Cas A, there are reasonably good historical records from the first British astronomer royal, Sir John Flamsteed, who observed this supernova in about 1680; its distance has been estimated to be 3 kpc [8]. However, as can be seen from Fig. 2, published values for the half-life of ^{44}Ti range from 39.0 to 66.6 yr [9–14]. Figure 3 illustrates the fact that this half-life range produces an uncertainty of a factor of 6 in the amount of ^{44}Ti ejected by the Cas A supernova. This uncertainty is much larger than that from theoretical estimates of the amount of ^{44}Ti produced in such a supernova event [3]. Because of the need for an accurate and reliable value of this important quantity, we performed two experiments to determine the half-life of ^{44}Ti. We presented a preliminary account of the first of these experiments last year [15]. Here we give the full details of both of them.

II. EXPERIMENTS

A. Source preparation and data acquisition

We produced ^{44}Ti via the ^{45}Sc$(p,2n)$ reaction using 40-MeV protons from the Lawrence Berkeley National Laboratory 88-Inch Cyclotron. This energy was chosen to be just above the maximum of the excitation function for this reaction as reported by McGee *et al.* [16]. However, in the course of preparing this source, we found far less ^{44}Ti than expected. We therefore remeasured the excitation function for this reaction and found that its maximum is at a much lower energy and has a significantly smaller peak cross section than previously reported [17]. To produce the source used in our first half-life experiment, a target of 99.9% pure metallic scandium 37.8 mg/cm^2 thick was irradiated for approximately 24 h with a proton beam of 1 μA. After allowing the short-lived activities to decay away, we chemically separated about 0.01 μCi of ^{44}Ti from the target. This activity was mixed with 0.04 μCi of ^{22}Na and 0.05 μCi of ^{137}Cs, and then dried and sealed in a small plastic planchet to form source 1. This *mixed* source together with a separate 1-μCi source of ^{241}Am was then rigidly mounted against the cap of a *dedicated* shielded 110-cm^3 high-purity germanium detector for γ-ray counting. Source 2 was produced approximately 2 yr later, by sealing together inside a small plastic box a thin metal foil of scandium and a thin metal foil of lead that had been irradiated several years earlier with 50-MeV proton beams. The lead foil contained approximately 0.04 μCi of ^{207}Bi. Following the completion of our first half-life experiment, source 1 was removed and the mixed ^{44}Ti/^{207}Bi source was then mounted against the cap of the same germanium detector. We collected γ-ray spectra of 4096 channels in 24-h intervals from source 1 for approximately 2 yr and recorded the data to magnetic disk using an ORTEC ACE data acquisition system on a PC. Then, using the same experimental setup, we collected data from source 2 for about 1 yr.

B. Data analysis

The activity of a radionuclide that has a half-life of 60 yr decreases by about 1% per year. Therefore, a *precise* deter-

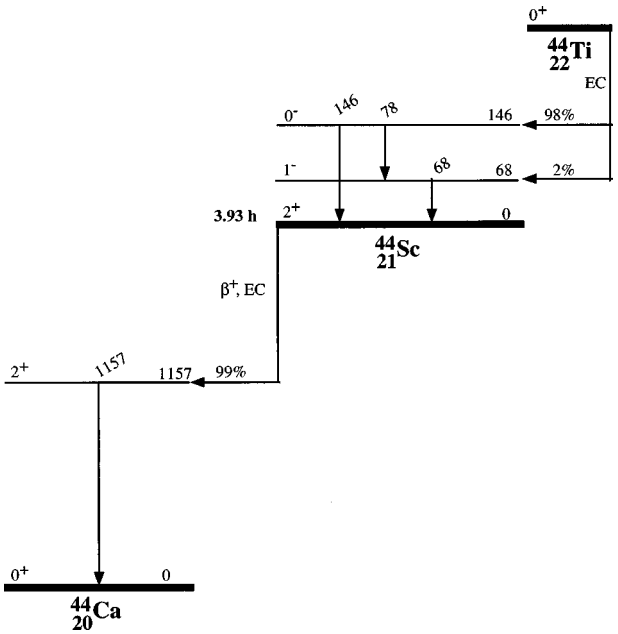

FIG. 1. Relevant portion of the decay schemes of ^{44}Ti and ^{44}Sc [1]. All energies are in keV. The 146-keV γ ray is very weak. Its intensity is approximately 0.1% of that of the 78-keV γ ray.

mination of spectral peak areas is needed to deduce its half-life if one measures the decrease in the γ-ray count rate for just a few years. Standard curve-fitting techniques for spectral peak-shape analysis usually provide accurate background subtraction for most uses in γ-ray spectroscopy; however, it is then difficult to estimate the size of the systematic error associated with this kind of background subtraction. At an early stage in the analysis of the data from this experiment, we compared the half-life values that we obtained by using the manual background subtraction procedure described below to those that resulted from the use of a sophisticated peak-fitting routine. The values obtained from these two different techniques agreed within their uncertainties. Therefore, in the analysis presented here we estimated the background for each peak of interest in the 10-day spectrum of source 1 by averaging the areas (for the same number of channels) above and below this peak. This procedure provided us a method for estimating the systematic error in the background subtraction, as will be discussed in the next section of this paper. For the 1157-keV peak (^{44}Ti) in source 2, we had to use a slightly modified procedure because coincidence sum peaks (1138 and 1149 keV) resulting from the internal conversion decay of the 570-keV level in ^{207}Pb contributed to the background in the lower-energy region of the 1157-keV peak. To determine the background we integrated the region of the spectrum below these sum peaks and interpolated this value to the region of interest under the 1157-keV peak.

We have determined the half-life of ^{44}Ti by fitting an exponentially decreasing function of time to the experimental γ-ray count rate using (with identical results) the following mathematical procedures.

(i) A linear least-squares fit to a logarithmic decreasing function of time, with the statistical weights appropriately modified. We changed the origin of coordinates to the center of gravity of the data so that the uncertainty in the slope

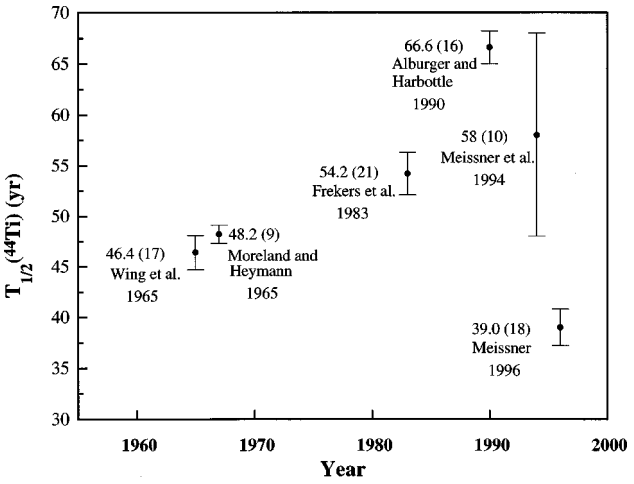

FIG. 2. Summary of previously reported values for the half-life of ^{44}Ti. Numbers in parentheses represent the 1σ uncertainties in the least significant digit(s).

(half-life) became independent of the covariance between this parameter and the intercept [18].

(ii) A nonlinear least-squares fit to an exponentially decreasing function of time. We used Bevington's [19] program subroutine CURFIT for this purpose.

C. Half-life determination from source 1

Figure 4 shows the sum of ten spectra, each measured for 1 day during the first 10 days after chemical separation of ^{44}Ti. The γ-ray peaks at 68 and 78 keV are from ^{44}Ti electron-capture decay, whereas that at 1157 keV is from the

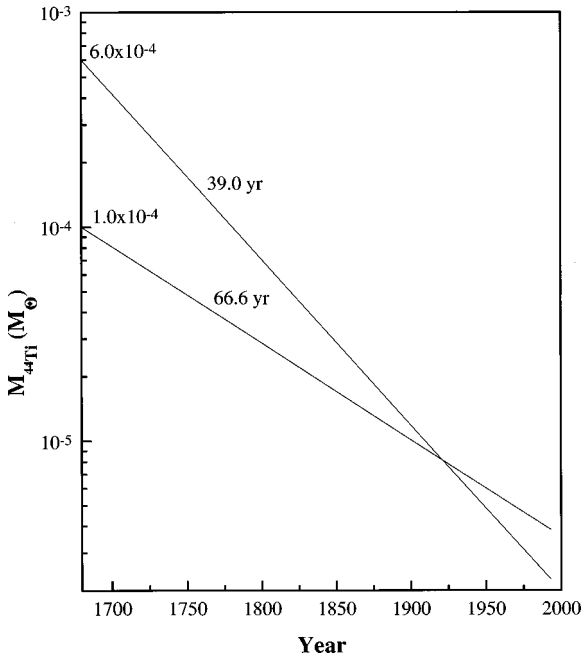

FIG. 3. Mass of ^{44}Ti (in units of solar mass) present in the Cas A supernova remnant versus time as inferred from the γ-ray observations of Iyudin *et al.* [4]. The two lines shown are labeled by the assumed half-life of ^{44}Ti. Notice that from the current γ-ray flux, the longer half-life value (66.6 yr) implies a *higher* present day ^{44}Ti abundance in this remnant, but a *lower* one at the time of the supernova explosion.

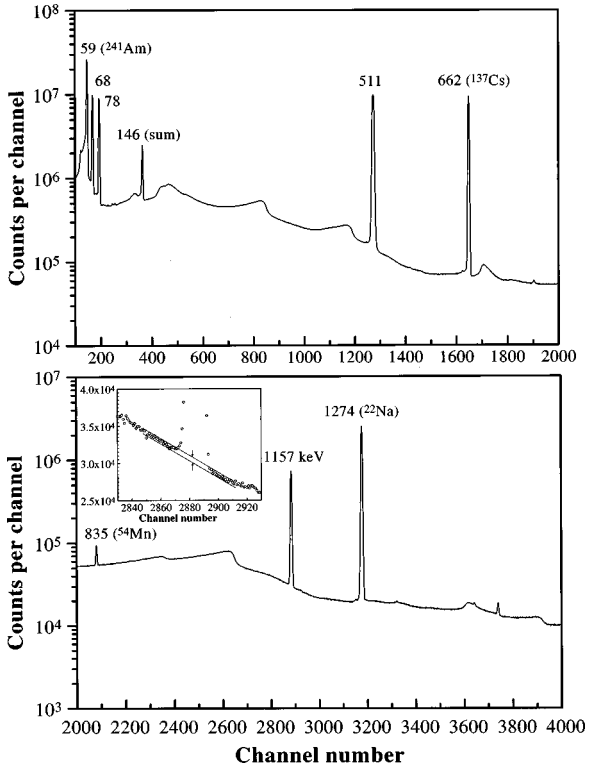

FIG. 4. γ-ray spectrum accumulated in 10 days of counting the mixed source of ^{44}Ti, ^{241}Am, ^{137}Cs, and ^{22}Na. All energies are in keV. Peaks labeled only by energy are from the decay of ^{44}Ti. The inset illustrates the background under the 1157-keV peak. The arrows indicate a $\pm 1\%$ systematic background uncertainty.

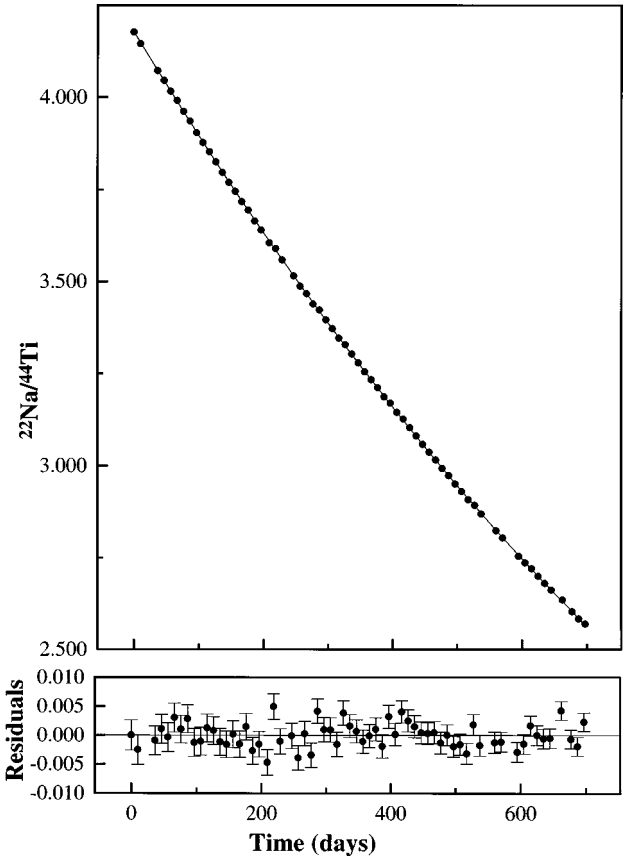

FIG. 5. The upper part of this figure shows the decrease in the ratio between the peak areas of the 1274-keV (^{22}Na) and 1157-keV (^{44}Ti) γ rays as a function of time. The curve going through the data is the result of a least-squares fit of an exponentially decreasing function of time. The ^{44}Ti half-life determined from this fit is 61.5(9) yr and $\chi^2/\nu = 1.1$. The lower panel shows the residuals to this fit.

decay of ^{44}Sc (3.9 h) in equilibrium with ^{44}Ti. The other peaks in the spectrum are from the standard radionuclides mentioned above and from a small ^{54}Mn impurity. Table I shows the counting statistics for the 1274-keV (^{22}Na) and 1157-keV (^{44}Ti) γ-ray peaks in the spectrum.

In order to correct the data for systematic errors that could have originated from variations in the detector performance or in the electronics over the long counting time required for this measurement, we fitted an exponentially decreasing function of time to the ratio between the peak areas of the 1274-keV (^{22}Na) and 1157-keV (^{44}Ti) γ rays. From fitting the decay rates of some γ rays from other radionuclides in our spectra, we found indications that small variations of this sort did occur during the course of our experiments. However, these ^{22}Na and ^{44}Ti γ rays have energies which are close to each other; consequently, the effect of detector instability would tend to cancel out in the ratio of their peak areas. The statistical uncertainty of about 0.06% in the ratio between the peak areas of these γ rays is sufficiently small to detect the decrease in the ^{44}Ti activity with time. Figure 5 shows the decrease in this count rate *ratio* with time, with a fitted effective decay constant $\lambda_{\text{eff}} = 0.255\,08(14)$ yr^{-1} and a χ^2/ν value of 1.1. Using our recommended value of $2.6024(12)$ yr^{-1} (see the Appendix) for the half-life of ^{22}Na [which corresponds to $\lambda = 0.266\,35(12)$ yr^{-1}], the decay constant of ^{44}Ti becomes

$$\lambda = 0.266\,35(12)\ \text{yr}^{-1} - 0.255\,08(14)\ \text{yr}^{-1}$$

$$= 0.011\,27(18)\ \text{yr}^{-1},$$

and its corresponding half-life is 61.5(10) yr. The number within parentheses represents the 1σ uncertainty in the least significant digit(s).

A possible systematic error could have originated from the background subtraction in the 1157-keV peak. The background under this peak is predominantly due to the Compton tail of the 1274-keV peak from ^{22}Na. Because of the much smaller background under the 1274-keV peak, the back-

TABLE I. Counting statistics for the 1274-keV (^{22}Na) and 1157-keV (^{44}Ti) γ-ray peaks in 10-day summed spectra taken at the beginning and at the end of the measurement.

	^{22}Na (1274-keV peak) Time		^{44}Ti (1157-keV peak) Time	
	$t=0$	$t=1.91$ yr	$t=0$	$t=1.91$ yr
Total counts	18.8×10^6	11.4×10^6	5.20×10^6	4.80×10^6
Background[a]	3.5%	3.8%	16%	11%
Net counts[b]	18.1×10^6	11.0×10^6	4.37×10^6	4.27×10^6
Uncertainty	0.024%	0.031%	0.056%	0.056%

[a]Percent of total counts.

[b]Total counts minus background.

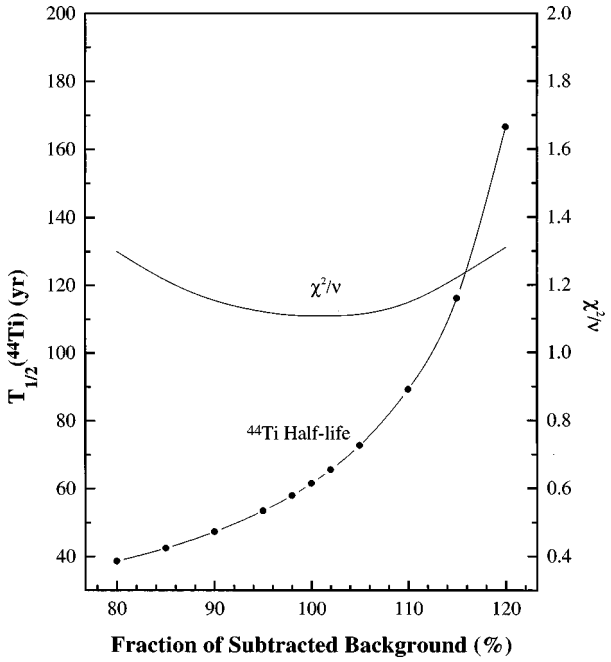

FIG. 6. Deduced half-life of ^{44}Ti and χ^2/ν as a function of the fraction of the background that was subtracted from the 1157-keV peak. Note that 100% corresponds to our best estimate of the actual background under this peak.

ground subtraction in this peak is not as significant a source of systematic error as is that for the 1157-keV line. The significant difference between the decay rates of the 1157- and 1274-keV γ rays makes both our measured values for

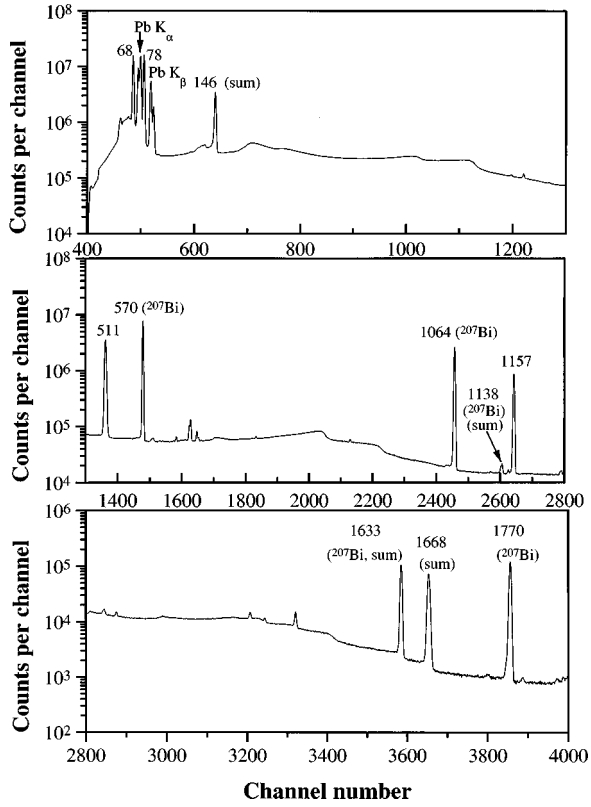

FIG. 7. γ-ray spectrum accumulated in 10 days of counting the mixed source of ^{44}Ti and ^{207}Bi. All energies are in keV. Peaks labeled only by energy are from the decay of ^{44}Ti.

TABLE II. Counting statistics for the 1064-keV (^{207}Bi) and 1157-keV (^{44}Ti) γ-ray peaks in 10-day summed spectra taken at the beginning and at the end of the measurement.

	^{207}Bi (1064-keV peak) Time		^{44}Ti (1157-keV peak) Time	
	$t=0$	$t=1.01$ yr	$t=0$	$t=1.01$ yr
Total counts	12.0×10^6	11.7×10^6	4.28×10^6	4.21×10^6
Background[a]	3.0%	3.0%	6.7%	6.6%
Net counts[b]	11.6×10^6	11.3×10^6	4.0×10^6	3.9×10^6
Uncertainty	0.030%	0.030%	0.052%	0.054%

[a]Percent of total counts.
[b]Total counts minus background.

the half-life of ^{44}Ti and the quantity χ^2/ν sensitive to the amount of background subtracted. To test this sensitivity, we varied the subtracted background under the 1157-keV peak about the value determined by using the procedure described in Sec. II B and then refitted the resulting decay curve. Figure 6 shows the dependence of the measured half-life and χ^2/ν on the subtracted background. From the shape of the spectrum in the vicinity of the 1157-keV peak and the shallow minimum of the χ^2/ν function around 100% of the suggested background, is possible to estimate a realistic background range of 99–101 % of the value we actually used. This range corresponds to a ^{44}Ti half-life between 60.0 and 63.5 yr.

D. Half-life determination from source 2

^{207}Bi may be conveniently used as a standard for measuring the half-life of ^{44}Ti. It has a half-life of about 33 yr and an intense 1064-keV γ ray (just 93 keV below the 1157-keV γ ray from ^{44}Ti). The half-life values of ^{207}Bi reported by various authors (see the Appendix), however, are not in good agreement with each other, and so our recommended value of 32.9(13) yr has a relatively large uncertainty of 4%. Figure 7 shows the sum of ten 1-day spectra. The small coincidence sum peaks between 1138 and 1149 keV are from true coincidences between the 1064-keV γ ray and Pb K x rays produced by internal conversions of the 570-keV transition. Table II shows the counting statistics for the 1064-keV (^{207}Bi) and 1157-keV (^{44}Ti) peaks in the spectrum.

Once again, in order to eliminate systematic errors, we fitted an exponentially decreasing function of time to the ratio between the 1064- and 1157-keV peak areas. Figure 8 shows the decrease in the γ-ray count rate *ratio* between the 1064- and 1157-keV peaks with time. From these data we have obtained an effective decay constant of $\lambda_{\text{eff}} = 0.009\,82(36)$ yr^{-1} and a value of 1.0 for χ^2/ν. As we have mentioned before, the effect of the detector instability on the measured half-life would tend to cancel out because the 1064- and 1157-keV γ rays are close in energy. Using our best value of 32.9(13) yr [which corresponds to a decay constant of $\lambda = 0.021\,07(83)$ yr^{-1}] for the half-life of ^{207}Bi, the decay constant of ^{44}Ti becomes

$$\lambda = 0.021\,07(83)\ \text{yr}^{-1} - 0.009\,82(36)\ \text{yr}^{-1} = 0.011\,25(90)\ \text{yr}^{-1},$$

and its corresponding half-life is 62(5) yr.

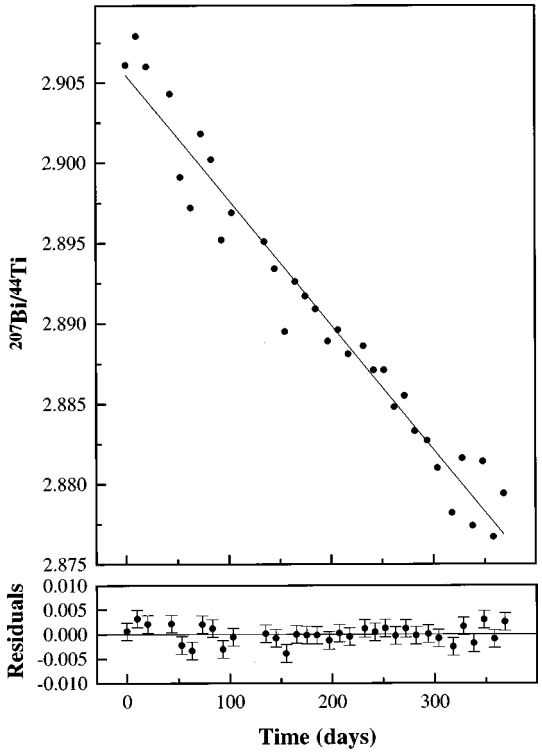

FIG. 8. The upper part of this figure shows the decrease in the ratio between the peak areas of the 1064-keV (^{207}Bi) and 1157-keV (^{44}Ti) γ rays as a function of time. The curve going through the data is the result of at least-squares fit of an exponentially decreasing function of time. The ^{44}Ti half-life determined from this fit is 62(5) yr and $\chi^2/\nu = 1.0$. The lower panel shows the residuals to this fit.

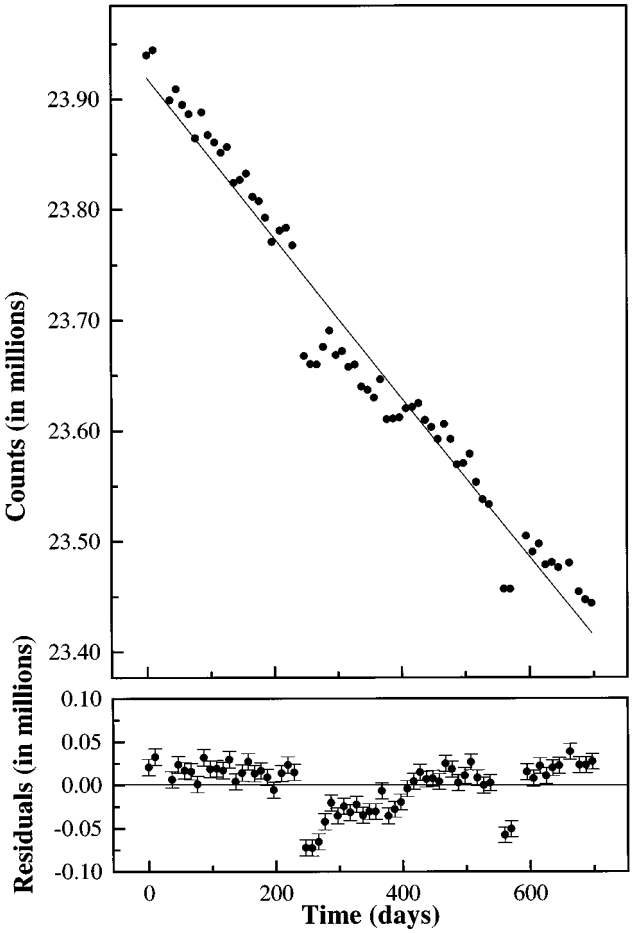

FIG. 9. The upper part of this figure shows the decrease in the count rate of the 78-keV ^{44}Ti γ ray as function of time deduced from Eq. (4). The dashed curve is the result of a least-squares fit of an exponentially decreasing function of time. The ^{44}Ti half-life determined from this fit is 62.3(5) yr and $\chi^2/\nu = 8.8$. The lower panel shows the residuals to this fit.

The background under the 1157-keV peak is mainly due to Compton tails from higher-energy γ rays from ^{207}Bi electron-capture decay. Since this radionuclide has a half-life that is just about half that of ^{44}Ti, the effect from a systematic uncertainty in the subtracted background is small. An estimated 5% error in the subtracted background would produce a ^{44}Ti half-life between 61 and 62 yr.

E. Verification of previous results

We described in Secs. II C and II D the use of ^{22}Na and ^{207}Bi as standards for measuring the half-life of ^{44}Ti. We present now a procedure to verify our results using *only* γ rays from ^{44}Ti electron-capture decay. It consists of measuring the decrease in the 78-keV γ-ray count rate with time and correcting this quantity for the instability of the detector photopeak efficiency. For the latter we used the peak area of the cascading 68-keV peak and also that of the 146-keV coincidence sum peak. This procedure uses the 68- and 146-keV peaks in each spectrum to determine the correct photopeak efficiency for the 78-keV γ ray at different times during the measurement.

The measured peak area of the 78-keV γ ray is given by

$$I_\gamma(78) = N_\gamma(78)\varepsilon(78)[1-\varepsilon_T(68)/(1+\alpha_{68})], \qquad (1)$$

where $N_\gamma(78)$ is the intensity of the 78-keV γ ray, $\varepsilon(78)$ is the detector *photopeak* efficiency for the 78-keV γ ray, $\varepsilon_T(68)$ is the *total* detector efficiency for the 68-keV γ ray, and α_{68} is the *total* conversion coefficient for the 68-keV transition.

Neglecting a 2% electron-capture feeding to the 68-keV level in ^{44}Sc, the measured peak area of the 68-keV γ ray becomes

$$I_\gamma(68) = N_\gamma(68)\varepsilon(68)[1-\varepsilon_T(78)/(1+\alpha_{78})] \qquad (2)$$

and that for the 146-keV coincidence sum peak,

$$I_\gamma(146) = N_\gamma(78)\varepsilon(78)\varepsilon(68)/(1+\alpha_{68}). \qquad (3)$$

Since the 68- and 78-keV γ rays have similar energies, we can use the same value for the ratio between their total and photopeak detector efficiencies, i.e., $\varepsilon(78)/\varepsilon_T(78) = \varepsilon(68)/\varepsilon_T(68)$. Combining Eqs. (1)–(3) and replacing $N_\gamma(78)/N_\gamma(68)$ by 1.06 [1], α_{78} by 0.031 [1], and $\varepsilon(78)/\varepsilon_T(78)$ by 0.84 (we measured a value of 0.829 for this ratio using the 88-keV gamma ray from a ^{109}Cd source), $N_\gamma(78)$ becomes proportional to

$$[0.86I_\gamma(68)/I_\gamma(146)+0.97][0.84I_\gamma(78)+I_\gamma(146)]. \qquad (4)$$

TABLE III. Summary of ^{44}Ti half-life results from the present experiments.

Measurement	Half-life (yr)	χ^2/ν	Systematic range
Source 1	61.5(10)	1.1	60.0–63.5 yr
Source 2	62(5)	1.0	61–62 yr
Verification	62.3(15)[a]	1.0	

[a]Actual value was 62.3(5) yr for $\chi^2/\nu = 8.8$.

Figure 9 shows a plot of $N_\gamma(78)$ given by Eq. (4) as a function of time. A least-squares fit of an exponentially decreasing function of time to these values yielded a ^{44}Ti half-life of 62.3(5) yr with a poor fit ($\chi^2/\nu = 8.8$). Although the statistical uncertainties in the areas of the 68-, 78-, and 146-keV peaks are similar to the uncertainty in that of the 1157-keV peak, possible systematic errors due to background subtraction may be significantly larger. Such errors are difficult to estimate because of the peak shape of these low-energy γ rays, and they may well be responsible for the quality of the fit. The half-life of ^{44}Ti deduced here, however, agrees with our other results and thus confirms the consistency of the data and our interpretation.

III. DISCUSSION OF RESULTS

Table III summarizes the results of our measurements for the half-life of ^{44}Ti. An average of the values in column 2 is 62 yr. Since these values may not be independent of each other, we have chosen the statistical uncertainty in the average half-life to be not smaller than the uncertainty in any individual measurement, that is, about 1 yr. If we combine this uncertainty with the systematic range of values due to our background subtraction shown in column 4, then our best value for the half-life of ^{44}Ti is 62 ± 2 yr.

A close value to our result is that of Alburger and Harbottle [12] (66.6 ± 1.6 yr). They measured the decay of a ^{44}Ti source for over 3 yr using a gas-flow proportional counter. Their system had a high detection efficiency and good stability, which they periodically tested with a ^{36}Cl source. The disadvantage of such a system, however, is the impossibility of energy discrimination. Therefore, the presence of a long-lived contaminant could have affected their results. Alburger and Harbottle searched for possible contaminants by measuring the γ-ray spectrum of their source and excluded (at a level above 1%) the existence of any γ ray (with energies below 3 MeV) from a contaminant. A pure beta minus emitter or one that emits weak γ rays, however, could have been present. Meissner [14] considered several beta-minus-emitting radionuclides as possible contaminants in the Alburger and Harbottle experiment. In particular Meissner identified ^{151}Sm ($t_{1/2} = 90$ yr) as a likely one. This radionuclide, which can be produced by the (n,γ) reaction with a thermal cross section of 104 b, emits a weak 21-keV γ ray that Alburger and Harbottle may have not detected. If 20% of the activity contained in their source originated from ^{151}Sm, their resulting half-life would have increased by about 4 yr, that is, from 62 to 66 yr. We have no explanation for the wide range of discrepant values of the half-life of ^{44}Ti reported by other authors.

TABLE IV. Reported values of the ^{22}Na and ^{207}Bi half-lives.

^{22}Na	
$T_{1/2}$ (yr)	Reference
2.6036(3)	Unterweger *et al.* (1992) [20]
2.6016(3)	Rutledge and Merritt (1980) [21]
2.6019(4)	Houtermans *et al.* (1980) [22]
	Best value 2.6024(12) yr
^{207}Bi	
$T_{1/2}$ (yr)	Reference
31.55(5)	Unterweger *et al.* (1992) [20]
32.7(8)	Lin and Harbottle (1991) [23]
34.9(2)	Alburger and Harbottle (1990) [12]
35.7(9)[a]	Yakonomura *et al.* (1978) [24]
38(4)	Rupnik (1972) [25]
38(3)	Appelman (1961) [26]
30.2(5)	Harbottle (1959) [27]
28(3)	Sosniak and Bell (1959) [28]
	Best value 32.9 (13) yr

[a]Revised value using the population of the 569.7-keV level in ^{207}Pb by ^{211}Po (0.516 s) decay $= 0.56$ (3)% [29] and $T_{1/2}(^{211}\mathrm{At}) = 7.214(7)$ h [32]. Half-life given in Ref. [24] was 33.4(8) yr.

As can be seen from Fig. 3, the longer half-life of ^{44}Ti deduced from the present experiment and that of Alburger and Harbottle [12] implies that the amount of ^{44}Ti required to account for the observed γ-ray flux from the Cas A supernova remnant is smaller than previously suggested. This smaller amount is easier to explain in the context of recent models of supernova nucleosynthesis [3]. It should also be pointed out that Ejnisman *et al.* [17] employed a value of 46 yr for the half-life of ^{44}Ti in order to deduce its production cross sections from the measured ^{44}Ti activity in their targets. In light of our present result for this half-life, these cross sections should be increased by a factor of $62/46 = 1.35$.

Note added in proof. After this paper was submitted, two other papers reporting new measurements of the ^{44}Ti half-life were submitted to Physical Review Letters [33, 34]. Their results agree with ours.

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APPENDIX

We have estimated best values for the half-lives of ^{22}Na and ^{207}Bi by averaging the experimental results listed in Table IV. The values given in each set are statistically inconsistent ($\chi^2/\nu \gg 1$). Therefore, we applied the *limitation of relative statistical weights* method [30], which has been specifically developed for averaging discrepant data and used before in the evaluation of standard radionuclides for detector calibration [31]. The weighted average for ^{22}Na is

2.6024(12) yr ($\chi^2/\nu=11$). Its uncertainty has been expanded to include the two most precisely measured values. The weighted average for ^{207}Bi is 32.9(13) yr ($\chi^2/\nu=29$). Here the small uncertainty of 0.05 yr given for the half-life in Ref. [20] was increased to 0.18 yr in order to reduce the statistical weight of this measurement from 92% to 50%. Again, the uncertainty in the weighted average has been expanded to include the most precise measured value of 31.55(5) yr [20].

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