Three-Laboratory Measurement of the ^{44}Ti Half-Life

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We report on a measurement of the ^{44}Ti half-life aimed at lowering existing systematic uncertainties in this value, which is important to astrophysics. The half-life was measured by following the decay of ^{44}Ti relative to ^{60}Co for about 5 yr and the measurements were performed independently in three laboratories—Argonne, Jerusalem, and Torino. We suggest to combine our result, 59.0 $\pm$ 0.6 yr, with the one from the accompanying Letter by Görres *et al.*, 60.3 $\pm$ 1.3 yr, to obtain a current "best value" for the half-life of ^{44}Ti of 59.2 $\pm$ 0.6 yr (1σ error). [S0031-9007(98)05567-7]

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The half-life of ^{44}Ti plays an important role in determining the amount of ^{44}Ti produced by cosmic rays in meteorites [1], and in supernova explosions [2–4]. ^{44}Ti production is currently determined via the 1157 keV γ line of the ^{44}Sc daughter in secular equilibrium (see Fig. 1). In order to determine the original ^{44}Ti production, the half-life value of ^{44}Ti enters twice: (i) in calculating the number of ^{44}Ti nuclei from the activity measurement, and (ii) in the correction for decay between production and measurement. Meteoritic ^{44}Ti measurements are usually performed for falls only, for which a date is known by direct observation of the fiery entry to Earth. Owing to the value of its half-life, ^{44}Ti produced in meteorites provides an important monitor of the galactic cosmic ray flux in the last few hundred years and its modulation by solar activity [1]. The first observation of the 1157 keV γ-ray flux from the Cassiopeia A supernova remnant, made by the space-borne γ-ray instruments COMPTEL [2,3] and OSSE [4], emphasized the necessity to solve the question of the uncertain ^{44}Ti half-life. Although further searches with COMPTEL on board of the Compton Gamma Ray Observatory did not reveal definite ^{44}Ti γ-ray signals from other supernova remnants [3], such signals may well be found with the increased sensitivity of future γ-ray observatories [e.g., the International Gamma-Ray Astrophysics Laboratory (INTEGRAL) to be launched [5] in 2001]. In calculating the total ^{44}Ti yield of a supernova from the ^{44}Ti activity observed in a space-borne instrument, uncertainties of four quantities (γ-ray flux, distance, explosion date, half-life) contribute to the overall precision of the ^{44}Ti yield result. Among these uncertainties, that of the half-life is still surprisingly large, despite the fact that it has been measured several times in the past (Table I). It can be seen in Table I that these half-life measurements for ^{44}Ti give discordant results. This situation, not uncommon for long half-life measurements, most likely results from the difficulty to reliably estimate systematic errors. This was the case, for example, for the important half-life of ^{14}C. Only after a reasonable understanding of systematic effects, a recommended half-life of 5730 $\pm$ 40 yr was finally accepted [12].

In view of the significance of the ^{44}Ti half-life, several attempts to remeasure it have been started [10,11,13]. In this Letter we present the results of a long-term γ-ray decay experiment of ^{44}Ti relative to ^{60}Co performed independently in three laboratories (Argonne National Laboratory, Hebrew University Jerusalem, and CNR Torino). The measurement of the decay of a radionuclide relative to another one is expected to minimize systematic errors arising from varying intrinsic and geometrical

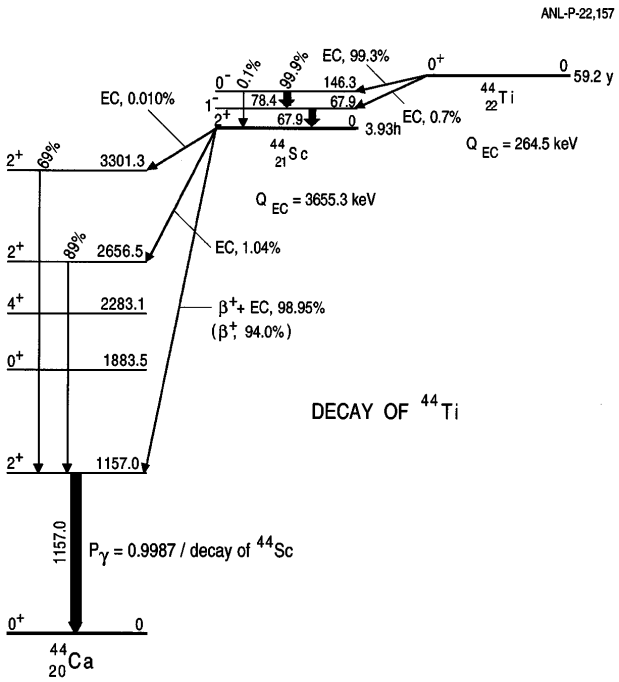

FIG. 1. Decay scheme of ^{44}Ti and its daughter ^{44}Sc.

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TABLE I. Summary of past ^{44}Ti half-life measurements.

Half-life[a] (years)	Method of measurement[b]	Reference
46.4 ± 1.7	Specific activity (MS + γ-act.)	Wing *et al.*, 1965 [6]
48.2 ± 0.9	Specific activity (MS + γ-act.)	Moreland and Heymann, 1965 [7]
54.2 ± 2.1	Specific activity (AMS + γ-act.)	Frekers *et al.*, 1983 [8]
66.6 ± 1.6[c]	Decay (β activity)	Alburger and Harbottle, 1990 [9]
58 ± 10	Specific activity (HISP + γ-act.)	Meissner *et al.*, 1995 [10]
63 ± 3	Decay (γ activity)	Norman *et al.*, 1997 [11]

[a]1σ uncertainties are given.
[b]MS = mass spectrometers, AMS = accelerator mass spectrometry, HISP = heavy ion spallation reaction with on-line counting of ^{44}Ti nuclei.
[c]2σ uncertainty is quoted in order to cover possible systematic effects [9].

efficiencies and from random summing effects. For this project ^{60}Co was chosen because of the closeness in energy of its 1173 keV γ line to the 1157 keV line in the ^{44}Ti decay and the precisely known value of its half-life [14], $t_{1/2} = 5.2714 \pm 0.0005$ yr. Three sets of sources were prepared at Argonne from the same source material and distributed to the three laboratories. Each laboratory performed independent decay measurements with these sources over several years.

A ^{44}Ti source of about 200 μCi was produced by the ^{45}Sc$(p, 2n)$ reaction using 45-MeV protons from the Juelich Isochronous Cyclotron. The details of the production and purification procedures have been described previously [8]. Using a solution of ^{44}Ti and a solution of ^{60}Co, three mixed sources of ^{44}Ti and ^{60}Co, and three pure ^{60}Co sources, were prepared. Each sample was prepared by placing a small drop of each solution in the center of a 1 in. diameter, 0.5 g/cm^2 thick Pt disk, drying them and then placing a piece of scotch tape to cover it. The Pt disk was placed in the recess of one-half of a Lucite disk, pressed by the other half of the disk, and the two pieces were then glued together with acetone. This produced a physically stable source. The Pt disk and the source location were clearly visible through the Lucite, showing no observable change over 5 yr. The three sets of sources had similar strengths: ~0.3 μCi ^{44}Ti+ ~0.3 μCi ^{60}Co for the mixed ones, and ~0.3 μCi for the pure ^{60}Co sources. The three mixed sources and the pure ^{60}Co sources were distributed to Argonne, Jerusalem, and Torino for the measurements. In addition, a pure ^{44}Ti source of similar strength as the mixed source was prepared for the measurement at Argonne. The spectra of the pure ^{60}Co and ^{44}Ti samples were measured with a shielded very-low-background Ge detector. No impurity γ-ray line was observed indicating that the ^{44}Ti and ^{60}Co sources were extremely pure.

At Argonne, the γ-ray spectra of the mixed ^{44}Ti + ^{60}Co and the two pure sources of ^{44}Ti and ^{60}Co, respectively, were measured with a 25% Ge detector, which had a resolution of 1.80 keV for the 1.3 MeV γ ray of ^{60}Co. No shield was built around the Ge detector because low background was expected above 1-MeV energy. A plastic holder with several slots was placed on the detector end cap, positioned, and held with plastic screws, such that the source-to-detector distance was 10.2 cm. The source in the Lucite disk was placed in an aluminum holder, taped, and then placed in the proper slot. The source was placed such that the source material was facing the Ge detector. Each sample was counted for a period of 48 h. In addition, a background spectrum with the source removed, but all the holder material in the same place was also taken. After measuring a set of four spectra, the source-to-detector distance was changed to 5.2 cm and again four spectra (three samples and background) were taken. These spectra were measured approximately every six months between February 1993 and June 1997, covering a period of 4 yr of decay. For the analysis, the respective background spectra were first subtracted from the sample spectra (Fig. 2). The counts in the 1157, 1173, and 1333 keV peaks were determined in several different ways. In one method, the background was determined by drawing a straight line between counts on the left side and right side of the peak and the background counts in the interval were subtracted from the counts in the peak of the spectrum. In another analysis, the background was taken as a step function. The analysis showed that half-life values determined using the peak counts by the various methods agreed within the error. In all cases, the areas of the three peaks in a given spectrum were determined in exactly the same way. Although the same Ge detector was used for all nine sets of spectra, the detector needed repair around the middle of the 4-yr period. After repair, the ratio of the 68 and 1157 keV γ lines of ^{44}Ti showed that the relative efficiency of the detector had slightly changed. Measurements also showed that the 1173/1333 ratio had changed by 0.16%. This correction was applied to the 1333 keV peaks measured after the Ge detector repair. The 1173 keV gamma ray is so close in energy to the 1157 keV γ ray that no correction was applied to this peak. In the Argonne measurement, the 1157/1173 and 1157/1333 ratios were obtained also from the spectra of the pure ^{60}Co and ^{44}Ti sources. This was done to eliminate the possibility that the ^{44}Ti(^{60}Co) peak areas in the mixed-source spectra may include background due to the ^{60}Co(^{44}Ti) lines. In both cases spectra

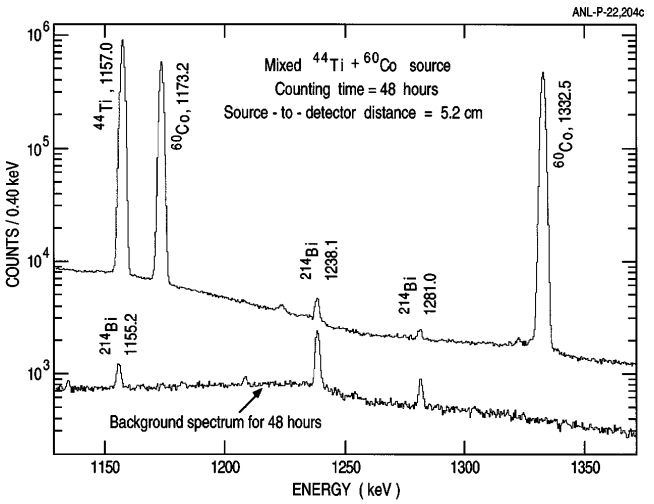

FIG. 2. A portion of the γ-ray spectrum of the mixed ^{44}Ti + ^{60}Co source (upper curve) and of the room background (lower curve) measured at Argonne with a 25% Ge detector, 3.0 yr after the half-life measurements were started.

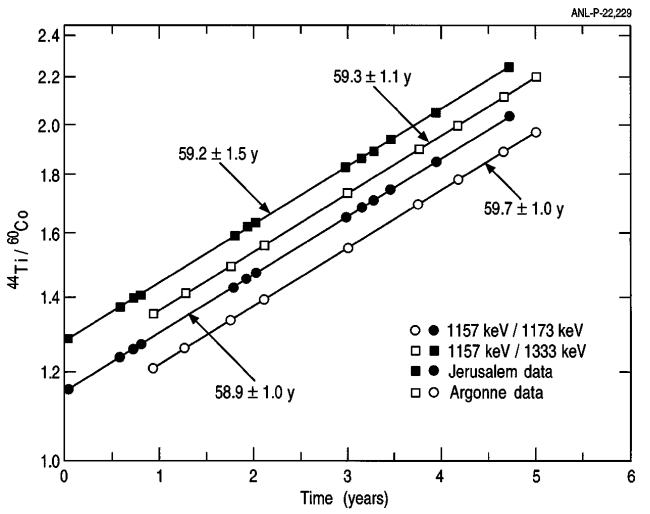

FIG. 3. Semilogarithmic plot of the ratios of the 1157 keV line in ^{44}Ti decay to the 1173 keV (circles) and the 1333 keV (squares) lines of ^{60}Co, measured as function of time in Jerusalem (solid symbols) and Argonne (open) from mixed ^{44}Ti + ^{60}Co sources. The ^{44}Ti half-life values determined from the slope (see text) of the lines are labeled. The figure shows two lines (measured at 5.2 cm distance) out of the eight measured at Argonne for the different combinations of mixed and pure sources and the two source-detector distances (see text).

measured at 10.2 and 5.2 cm were analyzed. In the measurement, the source holder was mounted on the detector just before the measurement began and was not permanently fixed to the detector. The source-to-detector distance thus was not constant for all the runs; however, once the source holder was fixed, the source-to-detector distance was constant for the three sources of ^{60}Co + ^{44}Ti, ^{60}Co, ^{44}Ti. Random summing corrections were applied in this case because the pure ^{44}Ti and ^{60}Co sources had different count rates in the Ge detector and the rate of change was different for both sources.

The ratios of the area in the 1157 keV peak to that of the 1173 keV peak as a function of time since the beginning of the experiment were fitted with an exponential function (Fig. 3). The slope of the line in a semilogarithmic plot directly gives the difference of the decay constants $\lambda(^{60}\mathrm{Co}) - \lambda(^{44}\mathrm{Ti})$. Using the known half-life of ^{60}Co, 5.2714 $\pm$ 0.0005 yr [14], the half-life of ^{44}Ti was deduced. The ^{44}Ti half-life was also determined from the 1157/1333 peak area ratios. The analysis was carried out both for the data measured at a source-to-detector distance of 5.2 and 10.2 cm. The fit of the data to an exponential decay was extremely good. In all cases, the 1σ error on the slope was between 0.2 and 0.4%. The weighted average of the four data sets of 1157/1173 ratios gave a value of 59.0 $\pm$ 0.8 yr for the ^{44}Ti half-life. A similar analysis of the four data sets of 1157/1333 ratios gave a value of 58.4 $\pm$ 1.3 yr. The latter value has a larger error due to the contribution of the uncertainty ($\pm$1 yr) in the efficiency ratio of the 1173 and 1333 keV γ rays. Because the ratios 1157/1173 and 1157/1333 are not truly independent of each other and the uncertainty in the latter is larger, we adopt 59.0 $\pm$ 0.8 yr as the final value of the Argonne measurement.

At Jerusalem, sets of spectra of the mixed source, pure ^{60}Co source, and background spectra were measured with a 35% Ge(Li) detector for a source-to-detector distance of 6.7 cm. The detector was not shielded from room background; a low-mass plastic holder in which the Lucite source disks could be positioned with high precision was kept mounted on the detector since the start of the measurements in July 1992, except for one interruption at $t = 1.6$ yr for detector service. No indication of change in the detector relative efficiency between the 1173 and 1333 keV lines of Co was observed.

Thirteen sets of spectra were measured during periods of 2 to 4 days over a 5-yr period. These spectra were analyzed with a peak fitting routine, using as model peak for the shape of the 1157 keV line of ^{44}Ti the experimental 1173 keV ^{60}Co peak measured in the same data set with the pure ^{60}Co source. A 60 keV region (identical for all data sets) around the 1157 and 1173 keV lines, measured with the mixed source, was thus fitted to the sum of two peak shapes scaled in energy and intensity and of a linear component representing background spectrum. The procedure gave excellent fits from which the ratio of the peak areas of the 1157 and 1173 keV lines was obtained directly from the ratio of the intensity scaling factors; the precision in this ratio, as determined by the fitting routine, was in all cases $\lesssim$0.2%. The ratios were corrected for the very small contribution ($\lesssim$0.06%) of the 1155 keV background line from ^{214}Bi, which sits under the 1157 keV line. In the same way as

was done at Argonne, the 1157/1173 intensity ratios as a function of time were fitted to an exponential function (Fig. 3). Using the value referenced above [14] for the ^{60}Co half-life, a value of 58.9 ± 1.0 yr (1σ error) was obtained for the half-life of ^{44}Ti. The ratios between the 1157 keV line in ^{44}Ti and 1333 keV line in ^{60}Co were also calculated. This was done using for the 1157 keV line the yield extracted from the model peak multiplied by the fitted intensity scaling factor of the 1157 line; the yields of the 1333 keV line were directly extracted from the mixed source spectra. The value of the ^{44}Ti half-life obtained from these ratios as function of time is 59.2 ± 1.5 yr, in agreement with the previous value. Here also, the final result of the Jerusalem measurement for the ^{44}Ti half-life is taken as the more precise value obtained from the 1157/1173 ratio, namely 58.9 ± 1.0 yr.

At Torino, two sets of spectra of the mixed source, separated by ~3 yr were measured due to the necessity, in between, to avoid interferences in the measurements of the very low activity of ^{44}Ti in meteorites performed by a Ge-NaI(Tl) gamma-spectrometer located close to the detector used for the mixed-source measurements. The detector used was a 25% HPGe detector placed in a very-low-background shielded cave. The source holder allowed the measurement of the two sets of spectra to be performed at precisely the same source-to-detector distance of 6.0 cm. Background counts under the peaks were subtracted using simple linear interpolation between adjacent regions around the peaks. The two ratios, 1157/1173 and 1157/1333, gave half-life values of 60.7 and 58.0 yr, respectively. The average of these values gave a half-life of 59.4 ± 1.4 yr (1σ error). The half-life was also determined from the absolute ^{44}Ti decay rates at the two measurement times, yielding a value of 59.1 yr in very good agreement with the relative decay value.

In summary, the values of the ^{44}Ti half-life measured at the three laboratories were: 59.0 ± 0.8 yr (Argonne), 58.9 ± 1.0 yr (Jerusalem), and 59.4 ± 1.4 yr (Torino). These values are in excellent agreement with each other, and a weighted mean gives the ^{44}Ti half-life as 59.0 ± 0.6 yr (1σ error). The confidence that this half-life is accurate within the uncertainty given, comes from the fact that it is the result of three independent measurements at three different laboratories. In addition, the Argonne value is the result of measurements at two different source-to-detector distances, and with an additional source (pure ^{44}Ti). It may therefore be reasonable to assume that systematic errors are largely averaged out.

Our half-life value agrees very well with that reported in the accompanying paper of Görres *et al.* [15], $t_{1/2}$ = 60.3 ± 1.3 yr. We feel that this agreement strongly supports the accuracy of both results and suspect that the disagreement with previous results (see Table I) is due to unaccounted systematic errors in these measurements, although we cannot rigorously prove this point. In this situation, it seems justified to obtain a "current best value" for the half-life of ^{44}Ti from the weighted mean of our result and that of Görres *et al.* [15], namely:

$$t_{1/2}(^{44}\mathrm{Ti}) = 59.2 \pm 0.6\ \mathrm{yr}, \quad (1)$$

where the quoted error is one standard deviation.

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