Long-term measurement of the half-life of ^{44}Ti

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We report on a series of precisely calibrated measurements of the 1157-keV gamma-ray emission rate from the decay of a single source of ^{44}Ti. These data were collected over a period of 20 years. We obtain a half-life of 60.7 ± 1.2 years. [S0556-2813(99)04801-3]

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The radioisotope ^{44}Ti is one of the few long-lived γ-ray-emitting isotopes expected to be produced in large quantity during a supernova explosion [1,2]. It decays slowly by electron-capture to ^{44}Sc, which subsequently decays with a half-life of 3.9 hours to the stable isotope ^{44}Ca, emitting a 1157-keV gamma ray 99% of the time. This 1157-keV gamma radiation has been recently observed in the supernova remnant Cassiopeia A, by the COMPTEL and OSSE instruments on board the orbiting Compton Gamma Ray Observatory [3–6]. The abundance of ^{44}Ti ejected by the supernova is of considerable theoretical interest [6]. This abundance can be calculated from the measured 1157-keV flux, the distance and time of the explosion, and the ^{44}Ti half-life. Until recently the half-life was the largest source of uncertainty in this calculation. ^{44}Ti is also found in meteorites, probably due to the spallation of cosmic rays on iron. The activity level can be used to indicate variations in cosmic ray flux and to calculate the terrestrial age of meteorites [7].

The laboratory measurement of half-lives of long-lived radioisotopes is notoriously difficult. There are two traditional methods: specific activity and exponential decay. In the first, the radionuclidic disintegrations per second and the quantity of radionuclidic atoms in a source are separately determined, and the ratio gives the decay constant and hence the half-life. These absolute measurements are often problematic and experimental uncertainties are difficult to estimate. In the second, the source must be counted by a reproducible detector system over a time long enough to see a measureable decay in the source activity. This is difficult to achieve when the half-life is long. Previously reported measurements of the ^{44}Ti half-life are shown in Table I. They disagree significantly, ranging from 39 years to 66 years. In 1997–98 the situation seemed to improve when four new results were reported that indicate a half-life of about 60 years [14–17]. These new measurements were all made relative to reference radioisotopes with well-known half-lives.

There is a long-standing program within the Ionizing Radiation Division of the National Institute of Standards and Technology to develop Standard Reference Material (SRM) radioactive sources. As part of this program we maintain germanium detectors with calibrated gamma-ray efficiencies up to 3 MeV. Over the years many measurements, using many different radioactive sources, have been carried out with these detectors. One such source in our possession is a sample of ^{44}Ti that was prepared in 1978. In response to recent interest in the ^{44}Ti half-life, we undertook to review these measurements, made over a period of 20 years, and use them for a half-life determination. Each measurement of interest was made using a well-calibrated detector in a known geometry, so a significant exponential decay of the source activity over this long period can be seen in these data, and a precise half-life can be extracted. This method is complementary to the other recent exponential decay measurements of ^{44}Ti, which were relative measurements made over much shorter periods.

The ^{44}Ti source was accelerator produced, and purchased from Amersham Inc. in 1978 as a 0.5 ml solution of $TiCl_4$ containing approximately 1.1×10^5 Bq of activity. A portion

TABLE I. Summary of previously reported measurements of the ^{44}Ti half-life.

Year	Authors	type	half-life (yr)	uncertainty (yr, 1σ)	Ref.
1965	Wing *et al.*	spec. act.	46.4	1.7	[8]
1965	Moreland *et al.*	spec. act.	48.2	0.9	[9]
1983	Frekers *et al.*	spec. act.	54.2	2.1	[10]
1990	Alburger and Harbottle	exp. decay	66.6	1.6	[11]
1995	Meissner *et al.*	spec. act.	58	10	[12]
1996	Meissner	spec. act.	39.0	1.8	[13]
1997	Norman *et al.*	exp. decay	63	3	[14]
1998	Norman *et al.*	exp. decay	62	2	[15]
1998	Ahmad *et al.*	exp. decay	59.0	0.6	[16]
1998	Görres *et al.*	spec. act.	60.3	1.3	[17]

TABLE II. Data summary.

Year	Days since Jan. 1, 1978	Detector	1157 keV γ emission rate (s^{-1})	Uncertainties (%) stat. (σ^{stat})	efficiency (σ^{eff})	source position (σ^{pos})	dead-time correction (σ^{dt})
1978	337	A	36861	0.182	0.93	0.23	0.2
1979	473	B	36770	0.180	0.60	0.18	0.2
1979	477	B	36762	0.127	0.60	0.18	0.2
1980	786	B	36271	0.268	0.60	0.18	0.2
1985	2751	C	33904	0.158	0.78	0.18	0.2
1995	6479	D	30300	0.280	0.28	0.13	0.2
1998	7703	D	29260	0.165	0.28	0.13	0.2

of this liquid was dried and sealed into a piece of polyester tape, forming an oval-shaped spot approximately 4.2 mm $\times$5.5 mm in extent. Visual inspection indicates that the integrity of the seal has remained intact and the spot of activity has not changed shape over the past twenty years. Also, there is no indication of radiation damage to the tape.

Four different gamma-ray detectors were used for these measurements. We will refer to these detectors by capital letters A–D. Detectors A and B are coaxial Ge(Li) crystals of volume 30 cm^3 and 60 cm^3, respectively. Their efficiencies from 120–2700 keV were measured using 24 single-radioisotope sources. An efficiency curve for each detector was obtained by fitting the logarithm of measured efficiencies to a power series in gamma-ray energy. Calibration lines in the vicinity of the 1157-keV gamma ray of ^{44}Ti include ^{46}Sc (1120.5 keV), ^{60}Co (1173.2 keV, 1332.5 keV), and ^{22}Na (1274.5 keV). More detailed descriptions of detectors A and B, and their efficiency calibrations, can be found in a previous publication [18]. Detector C is a 54 cm^3 coaxial *n*-type HPGe crystal; and detector D is an 81 cm^3 coaxial *p*-type HPGe crystal. The efficiency curves of these detectors were established using a number of NIST SRM sources, including ^{60}Co (1173.2 keV, 1332.5 keV) and ^{154}Eu (1004.8 keV, 1274.4 keV), and fit to a function of energy as described above. Descriptions of detectors C and D, and their efficiency calibrations, can also be found in previous publications [19,20].

Each detector was surrounded on five sides by a graded shield of copper, tin, and lead. The side facing the source was unshielded. The source-detector distance for detector A was 25 cm, measured using a precision spacer; and for detectors B–D was 25 cm, 10 cm, and 25 cm, respectively, measured optically using a telescope. Data were collected with computer-based MCA systems incorporating successive-approximation, fixed dead-time analog-to-digital converters (ADC's), using pulse shaping times of 4–6 μs. In each case the electronic dead-time correction was measured by feeding a 60 Hz pulser, at an equivalent energy of about 200 keV, into the detector preamp.

The 1157-keV gamma ray emission rate was determined using the following standard method, which is described in more detail in [21]. First, the 1157-keV peak is fit to a Gaussian plus a linear background to determine the peak position, background term coefficients, and full width at half maximum (FWHM). The background is then subtracted. The remainder is summed over a region of width 3.1 times the FWHM, centered on the peak position, to obtain the peak area. This is then divided by the efficiency to obtain the emission rate.

We have a total of six usable measurements of the ^{44}Ti source taken from 1978 to 1995, and we made an additional measurement in 1998. The data are summarized in Table II. Each data point represents several days of counting the source, and the tabulated time is the midpoint of the measurement. Figure 1 shows the relevant portion of the ^{44}Ti spectrum from the most recent measurement. The seven data points were fit to the exponential decay function:

$$A(t)=A_0 e^{-t/\tau}. \tag{1}$$

The dominant uncertainties in the data are the gamma-ray efficiency uncertainties. These are correlated for each set of points taken with a given detector because the same efficiency curve was used. All other uncertainties are uncorrelated. We obtained the best fit by minimizing the generalized chi-squared function that allows for correlated uncertainties (see for example [22]):

$$\chi^2=\sum_{i,j}\ (y_i^{\rm fit}-y_i^{\rm data})\,H_{ij}\,(y_j^{\rm fit}-y_j^{\rm data}). \tag{2}$$

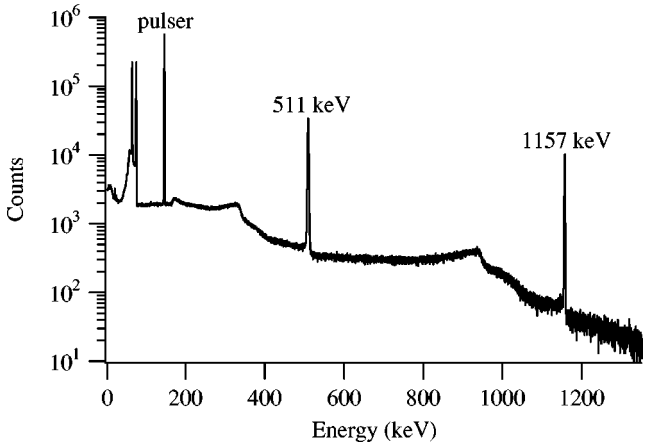

FIG. 1. ^{44}Ti spectrum from the most recent measurement, taken in 1998.

The matrix H is given by $H=C^{-1}$, where C is the covariance matrix, defined by

$$C_{ij}=\overline{(y_i-\bar{y}_i)(y_j-\bar{y}_j)}. \tag{3}$$

We constructed the covariance matrix using the data and fractional uncertainties listed in Table II as follows:

$$\text{diagonal terms:}\quad C_{ii}=y_i^2[(\sigma_i^{\text{stat}})^2+(\sigma_i^{\text{eff}})^2+(\sigma_i^{\text{pos}})^2+(\sigma_i^{\text{dt}})^2], \tag{4}$$

$$\text{off-diagonal terms:}\quad C_{ij}=y_iy_j(\sigma_{ij}^{\text{eff}})^2. \tag{5}$$

The off-diagonal terms are nonzero only for different points taken with the same detector. The best fit yielded $A_0=37185\ \text{s}^{-1}$, $\tau=31961$ days, and $\chi^2=2.13$ (5 degrees of freedom). This decay curve is plotted along with the data in Fig. 2.

The uncertainties in the fit parameters were found by the Monte Carlo method. Ten thousand sets of pseudodata were generated from Eq. (1), using the counting times from the actual data and the best-fit values of A_0 and τ. Statistical dispersion in the pseudodata were produced from the same covariance matrix, assuming Gaussian-distributed uncertainties. Each set of pseudodata was then fit to Eq. (1) as described above. This effort yielded histograms of A_0, τ, and χ^2 from which the standard deviations were calculated. We obtained $\sigma(A_0)=142\ \text{s}^{-1}$ and $\sigma(\tau)=658$ days. In addition, 77% of the pseudodata fits yielded $\chi^2>2.13$, so the quality of the fit to the actual data is good.

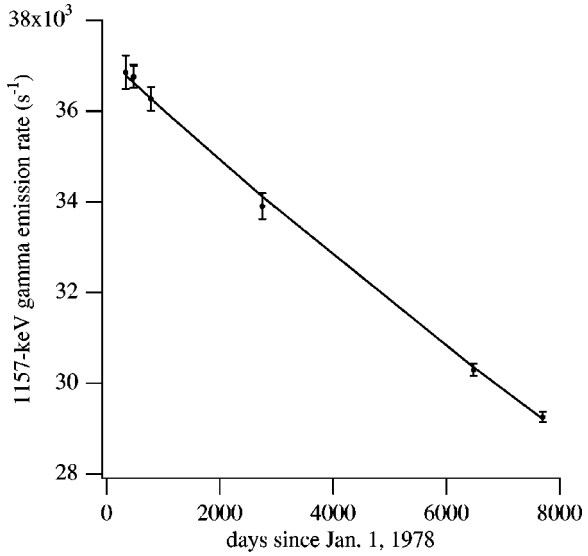

FIG. 2. Our data and the best-fit exponential decay curve.

From the analysis of our ^{44}Ti data we obtain a lifetime of 31961 ± 658 days, corresponding to a half-life of 60.7 ± 1.2 years (one standard deviation). This is in good agreement with the four most recent previous reports [14–17]. We emphasize that our result is an absolute measurement taken over a time period comparable to the half-life, so it is sensitive to different systematics than those of [14–17].

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