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HALF-LIFE DETERMINATION OF ^{41}Ca AND SOME OTHER RADIOISOTOPES

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ABSTRACT. We have performed a new determination of the half-life of ^{41}Ca by measuring the specific activity of an enriched Ca material with known ^{41}Ca abundance. We measured the activity *via* the 3.3-keV X-rays emitted in the electron capture decay of ^{41}Ca, and the ^{41}Ca abundance was measured by low-energy mass spectrometry. The result, $t_{1/2} = (1.01 \pm 0.10) \times 10^5$ yr, agrees with the recent 'geological' half-life of Klein *et al.*, (1991), $t_{1/2} = (1.03 \pm 0.07) \times 10^5$ yr, and with the corrected value of Mabuchi *et al.* (1974), $t_{1/2} = (1.13 \pm 0.12) \times 10^5$ yr. We recommend the weighted mean of these three measurements, $t_{1/2} = (1.04 \pm 0.05) \times 10^5$ yr, as the most probable half-life of ^{41}Ca. We also discuss the situation of the radioisotopes, ^{32}Si, ^{44}Ti, ^{79}Se and ^{126}Sn, whose half-lives, though still uncertain, are potentially interesting for future AMS studies and other applications.

INTRODUCTION

The half-life of a radioisotope can be determined principally in two ways. In the DECAY method, the temporal change of radioactivity is measured, either by radiation counting or by atom counting. For this, the absolute number of radioisotope atoms, N, need not be known. The half-life follows from the well-known exponential decay of radioactivity

$$N(t) = N(0)e^{-\lambda t}; \lambda = \ln 2/t_{1/2} \quad . \tag{1}$$

In the EQUILIBRIUM method, the radioactivity of a known number of radioisotope atoms is measured, at an arbitrary time, t. Here, the basic law of radioactivity applies

$$dN(t)/dt = -\lambda N(t) \quad . \tag{2}$$

For long half-lives, measurements in real time are always possible with the EQUILIBRIUM method, but often not possible with the DECAY method. A reliable geological time scale, however, allows the determination of very long half-lives by the DECAY method, such as demonstrated for 100 ka ^{41}Ca (Klein *et al.* 1991). Even though EQUILIBRIUM is measured mainly in real time, interesting possibilities exist for using steady-state conditions of cosmogenic radioisotopes, where, over geological time periods, production and decay have reached a secular equilibrium. To determine the half-life from this condition, one must know both the global production rate and the global inventory. Somayajulu *et al.* (1991, see below) recently estimated the ^{32}Si half-life in this way.

We first discuss the present status of the ^{41}Ca half-life, which has converged through the agreement of three independent measurements within the respective uncertainties. The ^{41}Ca case is similar to that of ^{14}C in 1962, when Godwin (1962) recommended a half-life of 5730 ± 40 years as the most probable value from the results of three concordant specific activity measurements.

We also discuss a few half-lives that are still uncertain. For ^{32}Si ($t_{1/2}$ = 100–172 yr) and ^{44}Ti (46–66 yr) several discordant results exist in the respective half-life ranges. Definite half-lives have not yet been established for ^{79}Se ($\leqq 6.5 \times 10^4$ yr) and ^{126}Sn (~1×10^5 yr). In general, the history of half-life measurements shows that only the agreement of several independent measurements can

establish the true half-life with certainty. Thus, the true value of a half-life where only one definite result exists may well lie outside the quoted uncertainty.

HALF-LIFE OF ^{41}Ca

The half-life of ^{41}Ca, which has been known for some time to be in the vicinity of 100 ka (Brown, Hanna & Yaffe 1953; Drouin & Yaffe 1962; Mabuchi *et al.* 1974), has gained renewed interest, because it became possible through accelerator mass spectrometry (AMS) to measure ^{41}Ca at natural concentrations. AMS measurements of ^{41}Ca in both extraterrestrial and terrestrial materials have been performed, with $^{41}Ca/Ca$ ratios in the range from 10^{-12} to 10^{-16}. Fink, Klein and Middleton (1990) summarized the various facets of this new field. Because of the long half-life of ^{41}Ca and its presence in bone, it was particularly intriguing to investigate the conditions for ^{41}Ca dating (Henning *et al.* 1987; Middleton *et al.* 1989; Kutschera *et al.* 1989a; Steinhof *et al.* 1989). Although the distribution of ^{41}Ca in terrestrial matter, including bones, appears to be complex (Kutschera 1990), and thus, a ^{41}Ca dating method has not yet been established, an accurate half-life of ^{41}Ca is ultimately desirable for any dating-related measurements of this radioisotope.

The decay of ^{41}Ca to ^{41}K proceeds through a pure electron capture (EC) transition. The available decay energy of 421 keV is below the electron-positron pair creation energy of 1022 keV, and thus, no positron emission is possible. Also, the decay populates only the ground state of ^{41}K. Thus, no discrete gamma rays accompany the decay. In the EC decay of ^{41}Ca, the electron is captured with 90% probability from the innermost orbit (K shell). This process is accompanied by a weak Inner-Bremsstrahlung continuum, with a spectrum extending up to the maximum decay energy (Fig. 1). The vacancy is filled with outer electrons, emitting with 14% probability (fluorescence

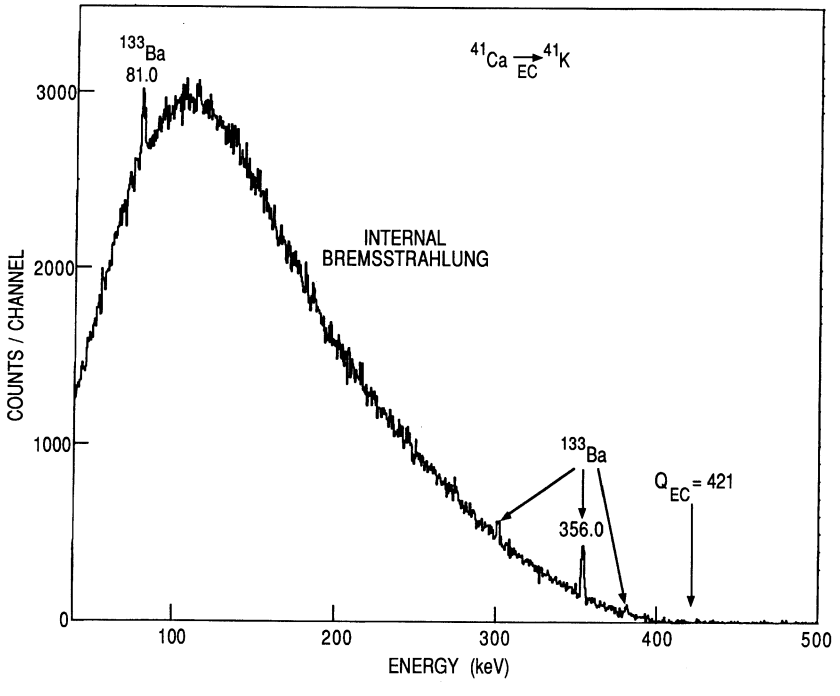

Fig. 1. Internal Bremsstrahlung (IB) emitted in the electron-capture decay of ^{41}Ca. The spectrum was measured in close geometry with a Ge detector from ~0.5 g of $CaCO_3$ containing ^{41}Ca with an isotopic abundance of 1.237%. We accumulated the spectrum for 30 min and subtracted a room-background spectrum. The observed spectrum does not represent the true IB shape, because it is strongly affected by the detector efficiency and absorption in materials between the source and the Ge crystal. We observe a low activity of 10.5-yr ^{133}Ba produced by the intense neutron irradiation of traces of barium in the calcium sample.

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yield) a soft potassium K X-ray of 3.3 keV. With higher probability (86%), the vacancy is filled by the Auger process, where electrons from outer orbits are ejected. These electrons are very difficult to detect for an element like Ca, which cannot be used in gaseous form in an ionization chamber. This leaves the 3.3-keV X-rays as the only useful radiation for an activity measurement. The almost radiationless decay of ^{41}Ca (the bulk of the decay energy is carried away by undetectable neutrinos), together with the long half-life and the low isotopic abundance, made it virtually impossible to detect ^{41}Ca in terrestrial materials prior to AMS.

At the previous ^{14}C conference, we gave a preliminary report (Kutschera *et al.* 1989a) on our effort to determine the half-life of ^{41}Ca by measuring the specific activity of an enriched Ca sample whose ^{41}Ca abundance had been accurately measured. We measured the specific activity with a Si(Li) detector *via* the 3.3-keV X-rays (Fig. 2). The abundance of ^{41}Ca (1.237%) was measured *via* low-energy mass spectrometry (LEMS) at Argonne (E. L. Callis, personal communication 1982) and at Caltech (D. A. Papanastassiou, personal communication 1988), with excellent agreement. The measurement of the specific activity of this material has now been completed, and Paul, Kutschera and Ahmad (1991) give a detailed account of these measurements elsewhere. Here, we summarize the essential features of our activity measurement, and compare it with other half-life results.

A reliable measurement of the specific activity of ^{41}Ca *via* the 3.3-keV X-rays has three necessary conditions:

1. The ^{41}Ca source must be thin enough to have negligible self-absorption.

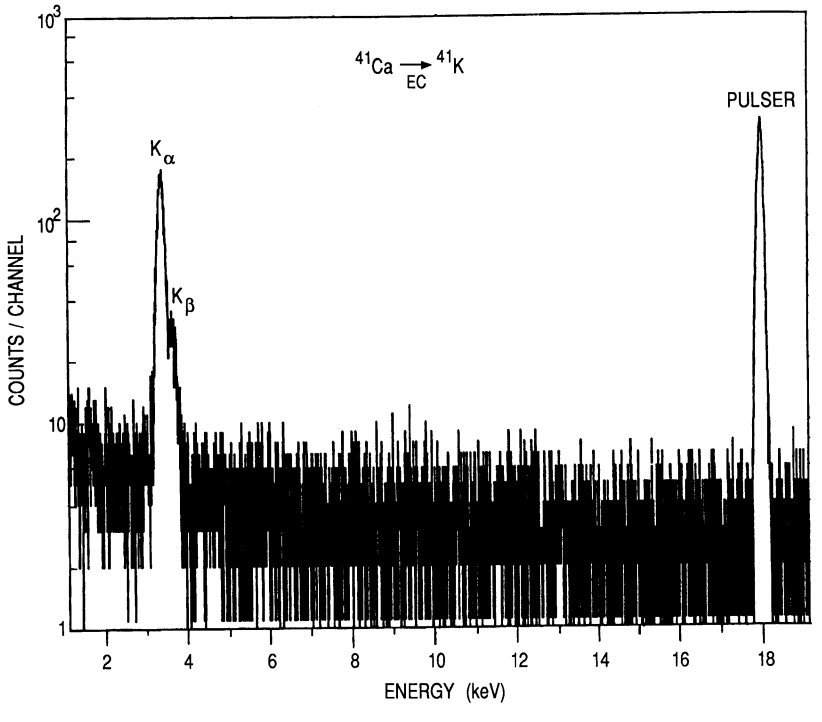

Fig. 2. Low-energy X-rays from a ^{41}Ca source, accumulated for 25.7 h with a Si(Li) detector. Clearly visible are the 3.3-keV X-rays from ^{41}K, following the electron-capture decay of ^{41}Ca. We used the pulser triggered with an external clock to determine the live time for data accumulation.

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2. The absolute efficiency of the Si(Li) detector for detecting 3.3-keV X-rays must be determined.
3. The probability of 3.3-keV X-ray emission per ^{41}Ca decay must be known.

We met the first condition by preparing ^{41}Ca sources of different thicknesses, and by observing no change in the specific 3.3-keV counting rate (cpm μg^{-1} Ca). We met the second condition by using different X-ray calibration sources (Fig. 3). Most important was the calibration with the 3.4-keV Np M X-rays from the decay of ^{241}Am, which lie very close in energy to the 3.3-keV X-rays of ^{41}K. Cohen (1988) has accurately determined the Np M X-ray intensities. Condition 3 requires the knowledge of both K-shell capture probability and fluorescence yield. Whereas the first quantity can be quite accurately calculated (Browne & Firestone 1986), the fluorescence yield is only known to about 5% accuracy (Krause 1979).

Combining the specific activity result with the ^{41}Ca abundance, we find a half-life of

$$t_{1/2}\ (^{41}Ca) = (1.01 \pm 0.10) \times 10^5 \text{ yr}.$$

In Figure 4, we compare our result with all previous half-life measurements of ^{41}Ca. Substantial corrections have been applied to the original half-life values of Brown, Hanna and Yaffe (1953) and Drouin and Yaffe (1962), which were changed from $(1.1 \pm 0.3) \times 10^5$ to $(2.0 \pm 0.6) \times 10^5$ yr, and from $(0.75 \pm 0.11) \times 10^5$ to $(1.6 \pm 0.3) \times 10^5$ yr, respectively. The corrections stem from changes in the adopted neutron capture cross-section of ^{40}Ca, and of the X-ray yield per ^{41}Ca decay. A smaller correction was necessary for the measurement of Mabuchi *et al.* (1974), which changed the half-life from $(1.03 \pm 0.04) \times 10^5$ yr to $(1.13 \pm 0.12) \times 10^5$ yr. In this particular case, however,

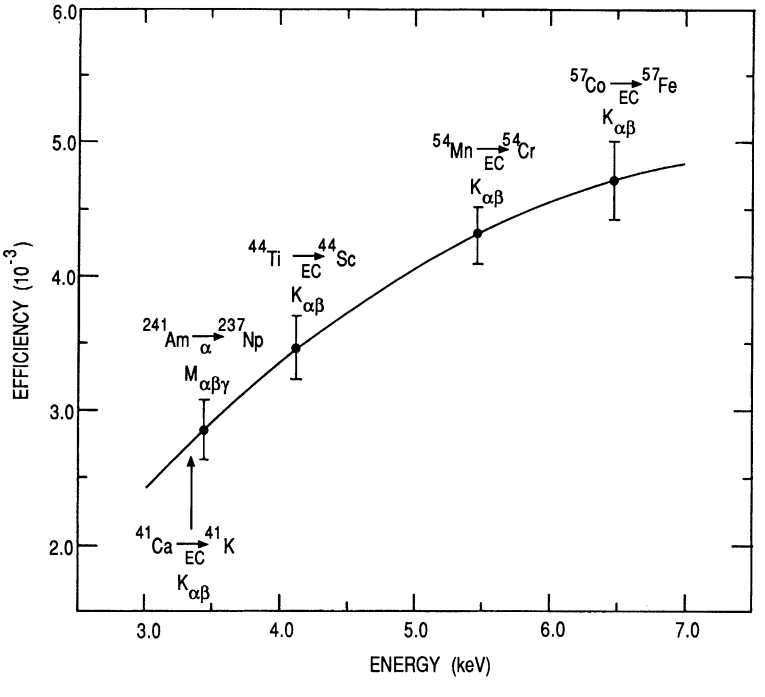

Fig. 3. Absolute efficiency calibration of the Si(Li) detector. The calibration points are labeled by the respective decay sequences. The solid curve is a parabolic fit to the four data points. From this fit, we obtained the efficiency for the ^{41}K $K_{\alpha\beta}$ X-rays.

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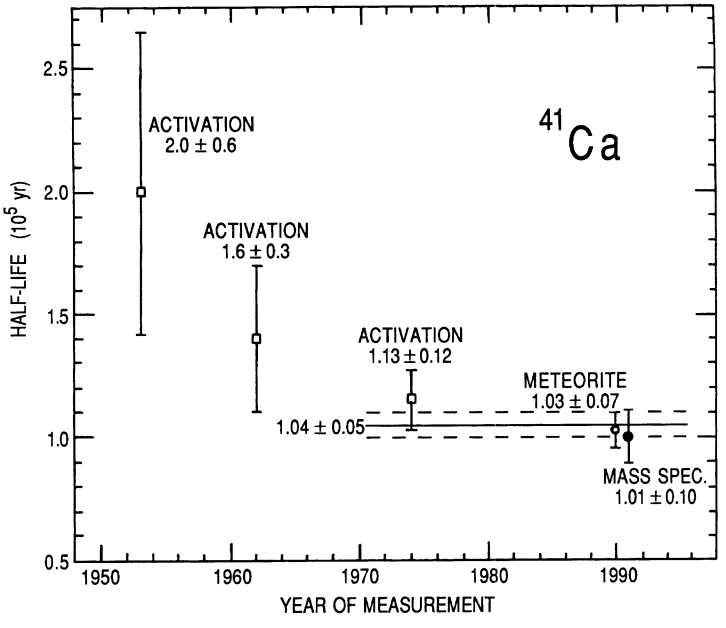

Fig. 4. Summary of results on half-life measurements of ^{41}Ca. The plotted values are labeled by the respective methods, and their half-life values in years. For the three measurements labeled with "activation," we give only the corrected values, which are due to changes in accepted neutron-capture cross-sections and fluorescence yields. For details, see Paul, Kutschera and Ahmad (1991). Also shown is the weighted mean, calculated from the three most recent half-life values (see text).

we increased the original uncertainty by a factor of three to make it consistent with the uncertainties inherent in the method. These three measurements, labeled "activation" in Figure 4, determined the ^{41}Ca abundance in the irradiated sample from the neutron-activation cross-section and the neutron exposure. This brings in the dependence of the half-life result on these two quantities. For our half-life measurement, we directly determined the ^{41}Ca abundance by mass spectrometry, which eliminates the uncertainties of the "activation" method.

Our result agrees well with the measurements of Mabuchi *et al.* (1974) and of Klein *et al.* (1991). Particularly striking is the excellent agreement of our half-life with the 'geological' half-life of Klein *et al.* (1991), which they determined from AMS measurements of the ^{41}Ca and ^{36}Cl content in Antarctic meteorites of different terrestrial ages. As a result of the three concordant half-life values, we feel encouraged to suggest that the weighted mean of these three measurements be accepted as the most probable half-life of ^{41}Ca

$$t_{1/2}\ (^{41}\mathrm{Ca}) = (1.04 \pm 0.05) \times 10^5\ \mathrm{yr}.$$

HALF-LIFE OF ^{32}Si

Half-life measurements have two classes of uncertainties. The first is the disagreement of different results outside their respective uncertainties. A notorious case of this kind is ^{32}Si, for which remarkable discordancies exist. The situation for this radioisotope is rather unusual, because DECAY and EQUILIBRIUM measurements in both real time and on a geological time scale have been performed. Figure 5 shows half-life results from these different measurements.

The three geophysical measurements involve different natural reservoirs. With the DECAY method, Clausen (1973) determined the half-life from the depth distribution of cosmogenic ^{32}Si in ice cores

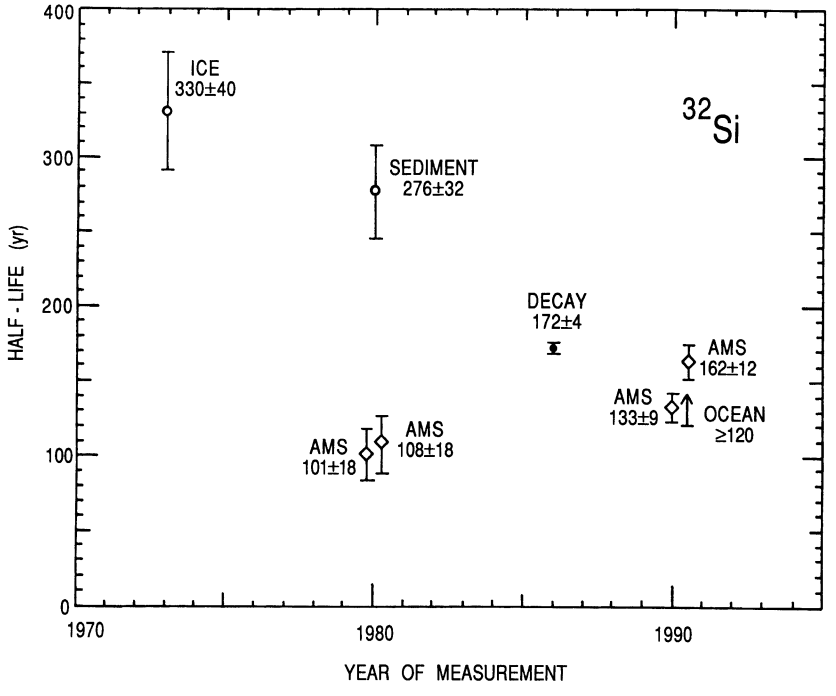

Fig. 5. Summary of half-life measurements of ^{32}Si. The results are labeled by the method or the respective natural reservoir, and by the measured half-life values in years. We give references to the measurements in the text.

from Greenland. DeMaster (1980) did a similar measurement in a sediment core from the Gulf of California. With the EQUILIBRIUM method, Somayajulu *et al.* (1991) determined a lower limit for the half-life from the comparison of the global ^{32}Si inventory of the oceans with the average cosmic-ray production rate. The ice-core result suffers from the limited knowledge of the temporal variation of both production and atmospheric precipitation of ^{32}Si (DeMaster 1980), which may reduce the half-life of Clausen (1973) considerably. Similarly, unvarved (unstratified) sections of the sediment core in the measurement of DeMaster (1980) may be due to 'slumping' (Cumming 1983), resulting also in a shorter half-life. The steady-state condition of cosmogenic ^{32}Si utilized by Somayajulu *et al.* (1991) compares two fairly well-established geophysical quantities. However, the unknown amount of ^{32}Si locked up in siliceous tests (shells) allows only a lower limit for the half-life (D. Lal, personal communication 1991).

Three of the four AMS measurements (Elmore *et al.* 1980; Kutschera *et al.* 1980; Hofmann *et al.* 1990) are performed with the EQUILIBRIUM method requiring absolute determinations of the $^{32}Si/Si$ isotopic ratio and of the $^{32}Si/Si$ specific activity. These measurements agree with each other within the relatively large uncertainties, although the agreement with the result of Hofmann *et al.* (1990) is marginal. However, all three results are clearly discordant with the geophysical results in ice and sediment, and also with the real-time DECAY measurement of Alburger, Harbottle and Norton (1986, see below). They are also discordant with the most recent AMS measurement of Thomsen *et al.* (1991), who utilized the ratio of two Si radioisotopes, $^{32}Si/^{31}Si$, to reduce the dependence on measuring absolute quantities. The main difficulty in this measurement is the relatively short half-life of ^{31}Si (2.6 h), requiring large decay corrections. However, the result apparently agrees with the only real-time DECAY measurement. Alburger, Harbottle and Norton (1986) obtained this latter result following the activity of a ^{32}Si sample over a period of four years,

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relative to a ^{36}Cl source, whose long half-life of 301 ka served as a time-independent calibration for a beta counter. In principle, such a decay experiment should give the most reliable result. On the other hand, the activity decreased by only 1.6% in four years, which imposes particularly stringent conditions on the precision of the activity measurement.

The final answer to the half-life of ^{32}Si awaits additional measurements. We know of one new variation of the EQUILIBRIUM method (Y. Chen *et al.* personal communication 1991), where ^{32}Si is produced in a fragmentation reaction of 2.6-GeV ^{40}Ar in a Be target. The ^{32}Si nuclei emerging with about 1.4 GeV from the target are separated in a mass spectrometer, and a known number of ^{32}Si atoms is collected. Measurement of the beta activity of the collector then allows the determination of the half-life. It will be interesting to see the result of this new measurement, although it may still not give the final answer because of the various discordant values shown in Figure 5.

HALF-LIFE OF ^{44}Ti

The situation of the ^{44}Ti half-life is similar to that of ^{32}Si, although fewer measurements are available (Fig. 6). Three EQUILIBRIUM measurements combine specific activity and mass spectrometry. Two of these measurements used LEMS (Wing *et al.* 1965; Moreland & Heymann 1965), and one used AMS (Frekers *et al.* 1983). The AMS result is in marginal agreement with the LEMS results. All three are, however, discordant with the real-time DECAY measurement of Alburger and Harbottle (1990). Thus, a new measurement is highly desirable.

An accurate half-life of ^{44}Ti is of particular interest in connection with supernova, SN1987A, since ^{44}Ti may be present in young remnants of supernovae (Clayton, Colgate & Fishman 1969). Gamma rays from 77-day ^{56}Co have already been observed from SN1987A (Matz *et al.* 1988). When the

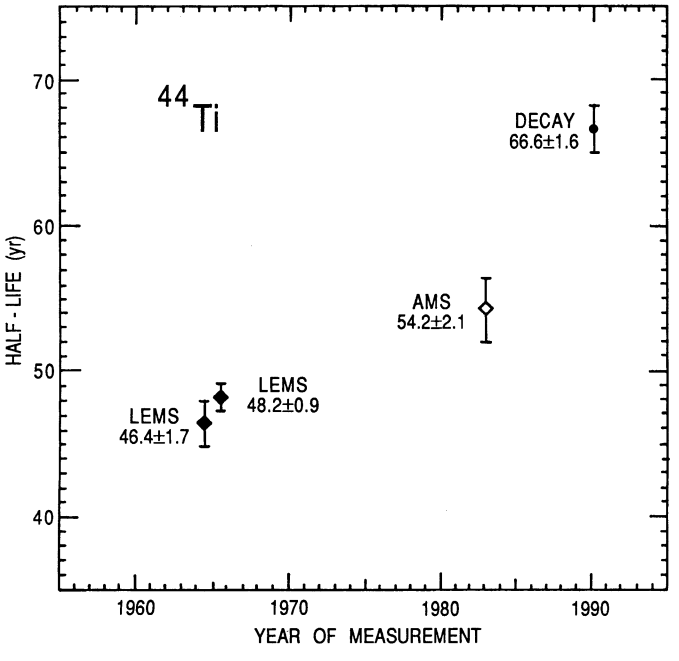

Fig. 6. Summary of half-life measurements of ^{44}Ti. The results are labeled by their method and the measured half-life values in years. We give references for the different measurements in the text.

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shorter-lived activities have died off, one hopes to find the 1157-keV gamma-ray activity of ^{44}Ti (Chevalier 1992). Knowing both the half-life and the time elapsed since the supernova explosion, one can calculate the initial amount of ^{44}Ti produced. ^{44}Ti is also of interest for tracing cosmic-ray production in the 100-year range in recent falls of meteorites (Bhandari *et al.* 1989).

We have started a project for a new real-time DECAY measurement, in order to establish the half-life of ^{44}Ti. We are preparing several gamma-ray sources from a master solution that contains approximately equal activities of ^{44}Ti and ^{60}Co. By following the decay over several years, we will measure relative intensity of the 1157.0-keV gamma line of ^{44}Ti and the 1173.2-keV gamma line of ^{60}Co in Ge detectors. With the well-known half-life of ^{60}Co, $t_{1/2} = 5.2719 \pm 0.0011$ yr (Walz & Weiss 1970), one should be able to measure accurately the half-life of ^{44}Ti from such a measurement. Because of the closeness of the two gamma-ray energies, possible small changes in the overall detector efficiency should negligibly affect the relative efficiencies of the two gamma lines. We plan to have sources measured at different laboratories to improve the overall reliability of the final half-life result.

Half-Life of ^{79}Se

Only an upper limit of $t_{1/2} \leqq 6.5 \times 10^4$ yr is known for ^{79}Se, from estimates of selenium activity observed in fission (Parker *et al.* 1949). A log ft >8.4 (where 1/ft is the energy-independent beta-decay transition probability), expected for the $7/2^+$ to $3/2^-$ beta transition from ^{79}Se to ^{79}Br, suggests a half-life longer than 1.5 ka (Singh & Viggars 1982). The beta-decay energy of 149 keV is similar to that of ^{14}C (156 keV). Since also the log ft value is similar, log ft(^{14}C) = 9.0, one may expect a ^{79}Se half-life similar to that of ^{14}C (5730 yr). The striking similarity of the decay characteristics of ^{79}Se and ^{14}C suggests a specific activity measurement of ^{79}Se with a ^{14}C beta-counting facility. This could be combined with an AMS measurement of a $^{79}Se/^{78}Se$ ratio measurement. Probably the best way to produce a measurable isotope ratio is neutron irradiation of ^{78}Se in a reactor.

Although we do not know of a specific application of ^{79}Se at this time, the probable proximity of its half-life to that of ^{14}C is intriguing. As a fission product of intermediate half-life, and with a fission yield comparable to ^{126}Sn (see below), it may be of similar or greater importance than the latter for nuclear-waste-related problems. With AMS, it also may be possible to detect ^{79}Se already in the environment. The long half-life also would be useful for AMS experiments of ^{79}Se in biomedical research, because of the negligible radiation exposure. Selenium is a very rare element in the human body, with a total amount of only about 20 mg (Snyder *et al.* 1975). It may be an important trace element, but its biological function is largely unknown. This situation is similar to that of aluminum, of which the human body contains about 300 mg. Meirav *et al.* (1990) and Barker *et al.* (1990) are currently investigating 720 ka ^{26}Al by AMS. Elmore *et al.* (1990) are also investigating 100 ka ^{41}Ca by AMS, in order to study the metabolism of the much more abundant calcium, of which the human body contains about 1 kg.

At this stage, ^{79}Se is a blank spot on the map of long-lived radioisotopes, which, by itself, makes it an interesting case, similar to ^{126}Sn described below.

Half-Life of ^{126}Sn

Like ^{79}Se, ^{126}Sn is a low-yield fission product, whose half-life was estimated from fission activity measurements to be in the 100 ka range (Dropesky & Orth 1962). Using the ATLAS facility at Argonne, we have attempted to measure this half-life by combining an AMS ratio measurement of $^{126}Sn/Sn$ (Kutschera *et al.* 1989b) with a measurement of the specific activity. The sample material was tin, extracted from a spent nuclear fuel rod. The activity measurement was quite

easily accomplished by measuring gamma rays from the decay sequence, $^{126}Sn \rightarrow {}^{126}Sb \rightarrow {}^{126}Te$, in a Ge detector. Figure 7 shows that, in the AMS measurement, we obtained a clean separation of ^{126}Sn from the stable isobar, ^{126}Te, using the gas-filled magnetic spectrograph (Paul *et al.* 1989). However, systematic uncertainties in the absolute $^{126}Sn/Sn$ ratio measurement prevented a reliable AMS result. Therefore, for the time being, this half-life also remains a blank spot on the map of long-lived radioisotopes.

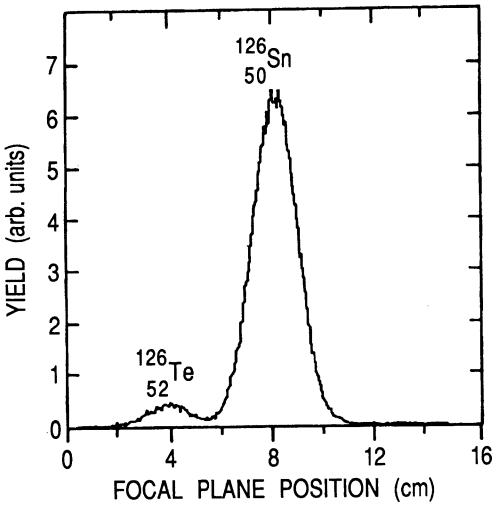

Fig. 7. AMS detection of ^{126}Sn in the gas-filled magnetic spectrograph from a sample with a $^{126}Sn/Sn$ ratio of about 10^{-7}. The separation from the stable isobar background of ^{126}Te is achieved with nitrogen gas of 6 torr pressure in the magnet, at an incident energy of 400 MeV from the ATLAS accelerator at Argonne (Kutschera *et al.* 1989b).

Interest in ^{126}Sn stems from its possible role as an important contributor to the overall radiation dose of high-level nuclear waste on a time scale of 0.01 to 10 ka (Patton & Penrose 1989). The measurements of ^{126}Sn in the marine environment (Koide & Goldberg 1985) and in sediments (Patton & Penrose 1989) *via* beta activity are disturbed by the presence of the 55-yr ^{121m}Sn activity. Clearly, AMS would easily separate these two isotopes. From our present experience of AMS measurements with ^{126}Sn, we can expect that $^{126}Sn/Sn$ ratio measurements should be possible with high sensitivity. However, a $^{126}Sn/Sn$ standard would be very desirable for such measurements, which brings up the question again of its half-life. With samples of low $^{126}Sn/Sn$ ratios ($\leqq 10^{-7}$), the development of a reliable AMS method to measure an absolute $^{126}Sn/Sn$ ratio is necessary, and has yet to be achieved. Samples with higher ratios in the range of $^{126}Sn/Sn \leqq 10^{-5}$ probably could be measured quite accurately with LEMS. The latter method looks more promising for establishing a half-life of ^{126}Sn.

SUMMARY

Long half-lives can be measured with a variety of methods, but often a period of discordant results precedes convergence toward true half-lives. We have presented one example (^{41}Ca) whose half-life is now well established. The other radioisotopes discussed in this paper show either discordant results (^{32}Si, ^{44}Ti), or only approximately known half-lives (^{79}Se, ^{126}Sn).

Several other radioisotopes' half-lives have been measured only once with a definite result (*i.e.*, with a quoted uncertainty). The history of half-life measurements suggests that several concordant results are needed before those half-lives can be accepted with confidence.

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