

New Measurement of the ^{60}Fe Half-Life

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We have made a new determination of the half-life of the radioactive isotope ^{60}Fe using high precision measurements of the number of ^{60}Fe atoms and their activity in a sample containing over 10^{15} ^{60}Fe atoms. Our new value for the half-life of ^{60}Fe is $(2.62 \pm 0.04) \times 10^6$ yr, significantly above the previously reported value of $(1.49 \pm 0.27) \times 10^6$ yr. Our new measurement for the lifetime of ^{60}Fe has significant implications for interpretations of galactic nucleosynthesis, for determinations of formation time scales of solids in the early Solar System, and for the interpretation of live ^{60}Fe measurements from supernova-ejecta deposits on Earth.

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Radioactive nuclei produced by astrophysical processes are a key for understanding the development of our Universe. The half-life of ^{60}Fe plays a prominent role in different astrophysical investigations. Most importantly, these are as follows.

Nucleosynthesis in the current Galaxy as observed through gamma rays.—Cosmic gamma rays detected [1] from the decay of its daughter nuclide ^{60}Co (1.17 and 1.33 MeV) are proof of the presence of ^{60}Fe in interstellar space and thus reflect ongoing nucleosynthesis in the Galaxy on the time scale of the order of 10^6 years. The inferred ^{60}Fe is believed to originate from massive stars, formed by successive neutron captures on preexisting Fe isotopes in neutron-rich regions inside these stars and ejected during the core collapse supernova. It has been argued that ^{26}Al and ^{60}Fe share at least some production sites in the Galaxy, as massive stars and supernovae are sources of both isotopes [2,3]. Ejection of significant amounts of ^{26}Al (but no ^{60}Fe) in the Wolf-Rayet phase preceding the supernova may blur this coincidence [4]. Recent γ -ray measurements by a space observatory [1] in conjunction with updated models for ^{26}Al and ^{60}Fe yields from massive stars of solar metallicity [3,5] were discussed with some controversy, though now in formal agreement (see, e.g., [6–8]). Note that the half-life of ^{60}Fe plays an essential role in such comparisons. Although the ^{60}Fe gamma-ray line flux under steady-state conditions does not depend on the half-life, with increased sensitivity individual ^{60}Fe source regions (such as the Cygnus region) will become observable. These, in general, are not in steady state; rather, their young, massive-star population evolves after birth according to stellar masses and their abundances. The γ -ray brightness from individual sources would be inversely proportional to the ^{60}Fe half-life, while the dependence of ^{60}Fe yields on the half-life is more complex due to different production and destruction rates within the dynamic stellar envelopes [3].

History of the early Solar System as observed through meteoritic inclusions.— ^{60}Fe decays via ^{60}Co to the stable nuclide ^{60}Ni . Solids condensed during the earliest phase of the Solar System preserve the amount of ^{60}Fe initially present in the interstellar cloud out of which our Solar System formed. The time from the collapse of this cloud until the formation of the planetary disk is in the range of 1×10^6 years. If a significant amount of ^{60}Fe survived until this stage, it acted as a heat source in melting and differentiation of early-formed planetesimals, such as found in meteorites [9]. Thus, any now-extinct ^{60}Fe determined as ^{60}Ni surplus in meteoritic inclusions serves as a sensitive chronometer for early Solar System history, i.e., possible nearby late-nucleosynthesis injection events, the evolution of the protoplanetary disk, and the proto-Sun. Recent measurements by Bizzarro *et al.* [10] show evidence for late supernova injection of ^{60}Fe into the protoplanetary disk, as also discussed by Wasserburg *et al.* [11]. They argue that the absence of ^{60}Fe in early-formed meteorites such as irons, pallasites, ureilites, and angrites could imply a late injection of ^{60}Fe into the disk, at a time when ^{26}Al was already homogeneously distributed in the inner Solar System. ^{60}Fe is exclusively of stellar origin, while ^{26}Al may also be produced in energetic collisions within the protosolar disk. Therefore, meteoritic ^{60}Fe abundances constrain the timing of its first appearance, its interstellar abundance, and also distribution within the protosolar disk, thus testing models of Solar System formation. An accurate knowledge of the half-life of ^{60}Fe plays a crucial role in the interpretation of such data.

Deposits of supernova ejecta on Earth as observed in ocean-crust material.—The finding of live ^{60}Fe on Earth was interpreted to be due to a nearby supernova that occurred several million years ago and deposited some of its ejecta on Earth [12]. Critical parameters in calculating the supernova distance are the expected amount of ^{60}Fe ejected by a supernova and how much of it de-

cayed before its measurement on Earth, a few million years later.

Two measurements of the half-life of ^{60}Fe had been reported until now. Roy and Kohman [13] obtained $t_{1/2} \approx 3 \times 10^5$ yr (uncertain by a factor of 3), and Kutschera *et al.* measured a value of $(1.49 \pm 0.27) \times 10^6$ yr [14], which is the currently accepted value used in the above interpretations. Obviously, a more accurate determination of the ^{60}Fe half-life is in great demand and might have significant impact on astrophysical data and their interpretation.

We determined the half-life $t_{1/2}$ by measuring three quantities in the same sample: the activity of ^{60}Fe , $A_{60\text{Fe}}$, the isotopic composition $N_{60\text{Fe}}/N_{\text{Fe}}$, and the number of iron atoms in the sample N_{Fe} .

The half-life is calculated from a modified equation of the radioactive decay

$$t_{1/2} = \frac{\ln(2)}{A_{60\text{Fe}}} \frac{N_{60\text{Fe}}}{N_{\text{Fe}}} N_{\text{Fe}}. \quad (1)$$

Compared to Ref. [14], about an order of magnitude more sample material was available, and the precise instrument of a multicollector inductively coupled plasma mass spectrometer (MC-ICP-MS) could be used to determine the number of iron atoms combined with γ -ray detection with a high-purity germanium (HPGe) detector for about 1000 days.

The ^{60}Fe nuclei had been extracted from a beam dump that was operated at the Paul Scherrer Institute (PSI) [15]. 3.6×10^{21} 590 MeV protons were stopped in this copper structure between 1980 and September 1992. After the dismantling of the facility, the copper beam dump was cut into pieces and stored. In 2004, when the short-lived radioactivities had decayed, the chemical separation of iron from the bulk material was performed.

The iron has been separated chemically from the bulk material as well as from the main contaminant ^{60}Co using liquid-liquid extraction and precipitation techniques. Details of the procedure can be found in Refs. [16,17]. 3.86 g Cu chips were used as starting material, and 5 mg stable iron and cobalt each were added as carrier. The final product was 5 mL of 0.1 M HCl solution containing the iron isotopes including ^{55}Fe with an activity of about 30 MBq. Its only detectable radiation (besides Mn x rays that are absorbed in the entrance window) is internal bremsstrahlung following the decay with a probability of 3.2×10^{-5} relative to K capture [18]. The total count rate in the detector never exceeded 115 Hz. The liquid was put into a glass vial (diameter 22 mm) and sealed (here called the *master sample*) [19].

The activity $A_{60\text{Fe}}$ was determined by the grow-in of the daughter isotope ^{60}Co via the two prominent γ -ray lines of 1.17 and 1.33 MeV. The decay scheme of ^{60}Fe is shown in Fig. 1.

Activity measurements started 50 days after the last separation step of cobalt. All measurements of the activity were done in a shallow underground laboratory with a

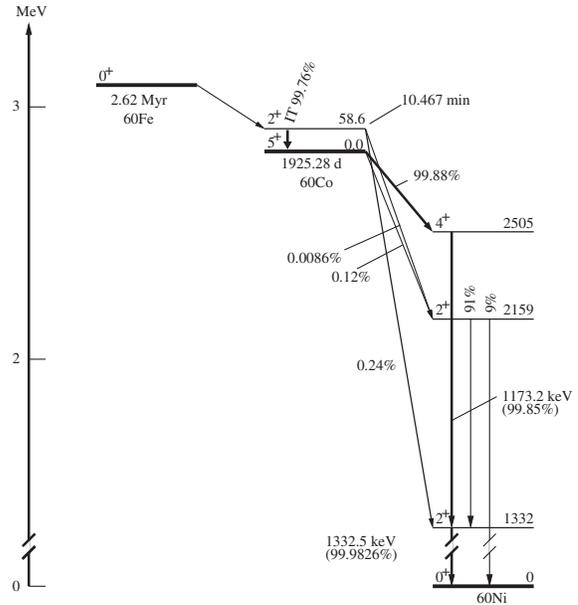


FIG. 1. Decay scheme of ^{60}Fe . All data are taken from Ref. [21], the data for $^{60}\text{Co}^m$ from Ref. [27], and the half-life value of ^{60}Fe from this Letter. The 1332.5 keV line is directly fed by $^{60}\text{Co}^m$; this leads to an increased probability in contrast to a ^{60}Co standard of 1.0025.

shielding of 15 mwe (meter water equivalent), to reduce cosmic ray induced background. The activity of the sample was monitored routinely by the two γ rays of ^{60}Co using a HPGe *n*-type detector with an active volume of 113 cm³ with a thin entrance window made of a 1.06 mm carbon fiber. The detector is housed in a lead shielding with a 5 mm copper lining and flushed with the boil-off nitrogen of the dewar.

In order to keep systematic uncertainties for the determination of the ^{60}Co - γ lines as low as possible, a ^{60}Co calibration standard in the same geometry as the iron sample was used. The initial activity of the calibration source in the weak acidic reference solution was 102.0 ± 1.5 Bq (1σ) [20]. This is in the range of the expected saturation activity of the iron sample. The background rate in the region of the ^{60}Co - γ lines was determined to be less than 10^{-5} cts/s/6 keV. The intensity of the two ^{60}Co - γ lines of our ^{60}Fe sample was easily measurable from the beginning of the measurement, and the initial ^{60}Co activity was found to be 0.2 Bq for the 1.17 MeV line. This corresponds to a decontamination factor of $\approx 10^7$. To avoid possible problems with a too close geometry, 233 days after the separation the sample was moved to 10 cm from the detector end cap. We measured the sample activity through the gradually increasing gamma-ray line intensity from decay of the daughter isotope ^{60}Co for almost 1000 days; see Fig. 2. The total count rate never exceeded 115 Hz in the beginning and 15 Hz after 233 days when changing the efficiency for the 1.17 MeV line of ^{60}Co from 1.16% to 0.156%. Most of the individual runs were performed for 10^6 s, resulting in more than 10^4 counts in

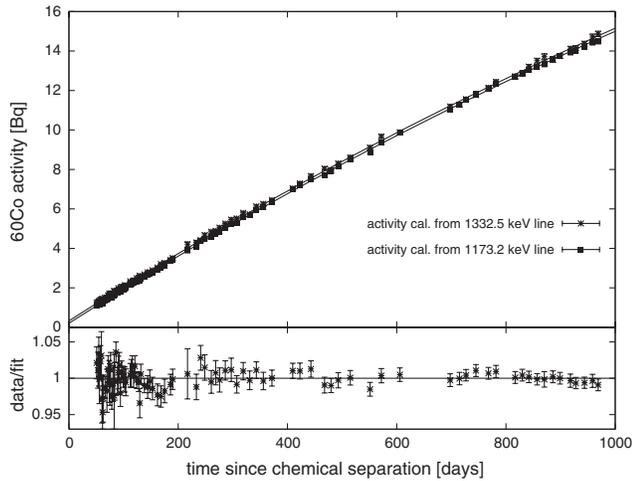


FIG. 2. Upper figure: Activity of the two ^{60}Co lines as a function of time. The solid curves are least-square fits of the function describing the feeding from the ^{60}Fe decay (see text). The upper curve corresponds to the 1332.5 keV line. The lower figure shows the data for the 1.17 MeV line normalized to the fit function.

the respective peak areas. The counting was paused for calibration runs with calibration runs at the end to confirm the detector performance.

The following equation was fitted to the measured activity for the 1.17 MeV line:

$$A_{60\text{Co}} = A_{60\text{Co}}^0 e^{-\lambda_{60\text{Co}} \cdot t} + A_{60\text{Fe}} (1 - e^{-\lambda_{60\text{Co}} \cdot t}) \quad (2)$$

and yielded a value of $A_{60\text{Fe}} = 49.14 \pm 0.08$ Bq and an initial $A_{60\text{Co}}^0 = 0.207 \pm 0.006$ Bq. In contrast to the standard, the 1.33 MeV line is also directly fed by $^{60}\text{Co}^m$ with a branching of 0.25%; see Fig. 1. A fit to this line gives a value of $A_{60\text{Fe}} = 49.25 \pm 0.08$ Bq. For the further calculation we combined both values to an $A_{60\text{Fe}}$ of 49.19 ± 0.11 Bq using the literature value for the half-life of ^{60}Co with $t_{1/2}(^{60}\text{Co}) = 1925.28 \pm 0.14$ d [21]. The error accommodates also uncertainties in $t_{1/2}(^{60}\text{Co})$ and the branchings.

The determination of N_{Fe} and $N_{60\text{Fe}}/N_{\text{Fe}}$ was performed by a MC-ICP-MS from the same master sample. Details of the system and the specific features are described elsewhere, e.g., Refs. [22,23]. Materials containing iron or nickel were avoided in the ion path. Additionally, a desolvating sample introduction system with a tenfold higher efficiency was used.

By weighing [24] the difference of the filled and the empty and dry glass vials, the weight of the master sample was determined to 4.545 ± 0.008 g.

For the determination of N_{Fe} , an aliquot of 100 mg was taken (here called the *ID sample*) from the master solution and diluted with 3% (w/w) nitric acid to approximately $4 \mu\text{g/g}$ for the measurement. Certified enriched material of ^{57}Fe [25] was prepared in the same manner to match the matrix and achieve similar signal intensities. The solutions

for isotope dilution mass spectrometry (IDMS) were prepared as 5 replicates with different spiking ratios from $^{57}\text{Fe}/^{56}\text{Fe} = 1.5$ to 0.75 to eliminate systematic errors.

For the ICP-MS measurement of the $N_{60\text{Fe}}/N_{\text{Fe}}$ ratio, an additional chemical treatment was necessary to determine the ^{60}Ni background at mass 60 correctly. It is important to note that the chemical efficiency of this purification does not influence the precision of the determination. Spallation reactions produce also stable nickel isotopes, causing a non-natural isotopic abundance. This was found experimentally and is also supported by Monte Carlo calculations. As a consequence, a nickel background correction—based on natural abundance—is not possible any more. In order to achieve natural isotope ratios for the nickel contaminants, an aliquot of 400 μL (here called the *N sample*) was used; 20 μg of Ni were added to the N sample, and Ni was then separated by precipitating $\text{Fe}(\text{OH})_3$ with ammonia solution. Ni remains in solution under these conditions forming a soluble Ni-ammonia complex. Repetition of this procedure several times gave a Ni-reduction sufficient for the background correction.

The measurements were performed relative to the certified reference material (CRM) IRMM-014 [26].

As the analysis procedure, the standard-sample-bracketing method was used. With this method potential drifts of the mass spectrometer can be detected, and the data can be corrected for these drifts. All data were corrected for background, mass bias, and isobaric interferences. The applied cup configurations are shown in Table I. The static procedure, i.e., solely line 1, was carried out for the CRM and IDMS measurements. For the N samples, a dynamic procedure (lines 1–3) using Faraday cups (FCs) and the secondary electron multiplier (SEM) was used. In this mode the electric and magnetic field of the mass spectrometer is altered to guide ions with a mass to charge ratio 57, 61, or 62 to the central detector; the detectors are not moved during the procedure. Furthermore, the central detector is switched from FC to SEM. The dynamic approach was necessary due to the very low Ni content which made it essential to apply the SEM instead of a FC to determine ^{61}Ni and ^{62}Ni in order to correct for the contribution of Ni to mass 60 on an individual basis for the 315 runs. The range of the correction of the signal was

TABLE I. FC configurations for the ^{60}Fe determination; for lines 2 and 3, a SEM was used at the central position. LX stands for a low position and HX for a high position; Int. time stands for the integration time for this configuration.

| Line | L3 | L2 | L1 | Center | H1 | H2 | H3 | Int. time |
|------|------------------|------------------|------------------|------------------|------------------|------------------|------------------|-----------|
| 1 | ^{54}Fe | ^{55}Fe | ^{56}Fe | ^{57}Fe | ^{58}Fe | ^{60}Fe | ^{61}Ni | 4 s |
| | | ^{55}Mn | | FC | ^{58}Ni | ^{60}Ni | | |
| 2 | ... | ... | ^{60}Fe | ^{61}Ni | ... | ... | ... | 4 s |
| | | | ^{60}Ni | SEM | | | | |
| 3 | ... | ^{60}Fe | ... | ^{62}Ni | ... | ... | ... | 4 s |
| | | ^{60}Ni | | SEM | | | | |

TABLE II. The various contributions to the uncertainty (1σ) of the three measurements are listed.

| | Rel. uncertainty [%] | |
|--|----------------------|-------|
| | Stat. | Syst. |
| $A_{60\text{Fe}}$ (master sample) | | |
| ^{60}Co standard | | 1.5 |
| Fit | 0.23 | |
| N_{Fe} (ID sample) | | |
| Weighing | | 0.18 |
| ID-ICP-MS | 0.28 | |
| $N_{60\text{Fe}}/N_{\text{Fe}}$ (N sample) | | |
| ICP-MS | 0.18 | |
| Total | 0.4 | 1.51 |

very stable and consistent for both isotopes and was about 15%.

The iron concentration in the ID sample was determined to $585.6 \pm 1.6 \mu\text{g/g}$, corresponding to 2.662 ± 0.009 mg of iron in the master sample with a non-natural abundance (atomic weight: 55.9022 ± 0.0033 g/mol).

The ratio $N_{60\text{Fe}}/N_{\text{Fe}}$ has been determined to be $(2.0483 \pm 0.0035) \times 10^{-4}$. An aliquot of the master sample gave a consistent result. The number of $N_{60\text{Fe}}$ in the master sample is therefore $(5.873 \pm 0.020) \times 10^{15}$.

The contributions to the uncertainty can be seen in Table II; the largest contribution originates from the ^{60}Co standard. A detailed account of these measurements will be published elsewhere. With Eq. (1) we obtain a half-life of ^{60}Fe of $(2.62 \pm 0.04) \times 10^6$ yr. This new value is much more precise and significantly higher than previously accepted.

The determination of an absolute amount of ^{60}Fe via accelerator mass spectrometry as in the previous determination [14] has been very challenging, and as outlined in Ref. [14] there has been the potential of a loss of ^{60}Fe counts at the measurement resulting in a too short half-life.

The understanding of the early Solar history needs the knowledge of a precise half-life of ^{60}Fe . There is no conclusive model at present which can explain the presence of ^{60}Fe in the early Solar System in a satisfying way. Since our determination of the half-life yields a 1.76 higher value, any models have to be reviewed. Also, with our present result the error in the half-life of ^{60}Fe contributes only a little (about 1.5%) to the uncertainties of these models.

Furthermore, a supernova deposit of ^{60}Fe on Earth [12], assumed some 2 to 3 Myr ago, should now yield a lower value at that time because of the longer half-life. Hence a more distant source must be assumed.

Also, future detection of ^{60}Fe in individual γ -ray sources needs a precise half-life of ^{60}Fe . Any uncertainty is then dominated by experimental errors of the γ flux and distance measurements. Our result for the ^{60}Fe half-life will

then provide considerably more stringent tests of supernova models than presently exist.

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