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Quantification of ^{60}Fe atoms by MC-ICP-MS for the redetermination of the half-life

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Abstract In many scientific fields, the half-life of radionuclides plays an important role. The accurate knowledge of this parameter has direct impact on, e.g., age determination of archeological artifacts and of the elemental synthesis in the universe. In order to derive the half-life of a long-lived radionuclide, the activity and the absolute number of atoms have to be analyzed. Whereas conventional radiation measurement methods are typically applied for activity determinations, the latter can be determined with high accuracy by mass spectrometric techniques. Over the past years, the half-lives of several radionuclides have been specified by means of multiple-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) complementary to the earlier reported values mainly derived by accelerator mass spectrometry. The present paper discusses all critical aspects (amount of material, radiochemical sample preparation, interference correction, isotope dilution mass spectrometry, calculation of measurement uncertainty) for a precise analysis of the number of atoms by MC-ICP-MS exemplified for the recently published half-life determination of ^{60}Fe (Rugel et al, Phys Rev Lett 103:072502, 2009).

Keywords Multiple-collector inductively coupled plasma mass spectrometry · Half-life · Isotope dilution

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Introduction

The half-life of a radionuclide represents an important nuclear physics property. A precise knowledge of this quantity is a precondition in various basic scientific fields like nuclear physics and nuclear astrophysics as well as in applied research areas, for example, geosciences, climate reconstruction, archeology, cosmology, life science, and many others. Investigations in nuclear astrophysics are aimed to enlarge our understanding of the origin and the development of the universe, studying the production and decay of isotopes in the nuclear synthesis and, thus, explaining the natural abundances of stable isotopes. In particular, long-lived radionuclides that are formed in these nuclear processes play an essential role as so-called "waiting points," allowing for several follow-up reaction paths. Unfortunately, in many cases, the literature values of long-lived radionuclides differ widely and their precisions also vary, since the methods of determination and often also the amount of available pure material are limiting factors. In nuclear astrophysics, the half-life values influence the calculation of production and decay rates of elements in the universe as well as the explanation of isotopic ratios of stable elements. Another, more applied example is accelerator mass spectrometry: unknown radioactive samples are measured with respect to reference standard samples. Their standard value is defined by a given activity and half-life. Therefore, all measurements imply systematic errors depending on the accuracy of the used half-life value. This is of special importance, if such measurements are used for dating, for instance, for climate reconstruction [2] or for age determination of archeological artifacts [3]. Furthermore, the interest in more accurate half-life values of cosmogenic radionuclides was growing, and accordingly, the demand for suitable sample material increased.

While half-lives of minutes up to some years can be determined with high precision by measuring the activity of the isotope over time, the task gets more and more challenging with increasing half-life. For radionuclides with half-lives in the million-year range, the direct decay measurement is not applicable. The only alternative method is to determine the half-life $t_{1/2}$ by measuring the ratio of the activity A of a radionuclide and the number of atoms N via the following equation:

$$t_{1/2} = \ln(2) \cdot \frac{N}{A} \tag{1}$$

Accurate activity determinations can be performed using conventional radiation measurement methods, dependent on the decay properties of the radionuclide. Mass spectrometry, especially multiple-collector inductively coupled plasma mass spectrometry (MC-ICP-MS), has proven to be a valid technique in the recent past for the determination of number of atoms N [4–7]. The high accuracy of the method, combined with the large number of accessible elements makes MC-ICP-MS ideal for half-life determinations. Other techniques, such as thermal ionization mass spectrometry (TIMS), are superior in terms of accuracy. But, TIMS suffers from low ion yield for elements with high first ionization potential and provides no access to a number of elements, e.g., W and Hf.

^{60}Fe has a special status among the exotic radionuclides, as it is of great importance for several astrophysical topics like the element synthesis in the galaxy as observed through gamma rays [8], the history of the early solar system traced by meteoritic inclusions [9], and deposits of supernova ejecta on earth as indicated in ocean-crust material [10]. Unfortunately, the possibilities of man-made production of ^{60}Fe to gain sufficient material for experimental investigations are limited. The possible double neutron capture of stable ^{58}Fe requires extremely high neutron fluxes, which are only available in certain reactors. In addition to the high flux requirements, very long exposure times lead to time-consuming and expensive procedures. In 2009, the consequence was that basic nuclear data of ^{60}Fe were either not very precise, like the half-life (only two measurements have been performed till that time [11, 12]) or completely missing, like the neutron capture cross sections of ^{60}Fe, which could be determined using a new sample material from a source described below [13].

An efficient alternative approach is spallation reactions, induced by highly energetic projectiles on targets with higher atomic number—a method which depends on the availability of high-power accelerators. Highly activated materials like targets, beam dumps, and shielding material from those accelerators may contain considerable amounts of desired radionuclides like ^{60}Fe. Therefore, some years ago, an initiative was started at Paul Scherrer Institute (PSI)—operating the worldwide most powerful 590 MeV ring cyclotron with a proton beam current of up to 2.4 mA [14]—that aims to exploit accelerator waste as a source for rare and exotic isotopes using chemical separation methods (project Exotic Radionuclides from Accelerator Waste for Science and Technology). One of the top priority isotopes of this program is ^{60}Fe; some 10^{16} atoms could already be separated [15]. Further activities are ^{44}Ti, ^{26}Al, ^{53}Mn [16], and ^{10}Be [17].

With a sample containing about 6×10^{15} atoms ^{60}Fe, a remeasurement of the half-life value was carried out using γ-spectrometry for the activity determination and MC-ICP-MS to measure the number of atoms [1]. The present paper aims to provide an elaborate description of the MC-ICP-MS procedure used for the redetermination of the ^{60}Fe half-life.

Experimental description

Sample preparation

The first critical aspect to fulfill is the isolation of the nuclide of interest in significant amounts and—in an ideal case—free of isobaric interferences. The source of the ^{60}Fe sample was a cylindrical copper beam dump from the former Bio Medical Area of PSI. This beam dump was exposed to about 0.1 Ah of highly energetic protons (590 MeV). After the shutdown of the facility in 1992 and a cooling time of almost 12 years, the beam dump was subjected to an extended sampling to study the radionuclide inventory distribution by radiochemical separation techniques [18]. Thereafter, the central part of the beam dump, where roughly 80 % of the total activity was concentrated, was drilled out. In total, 3.86 g of the produced copper chips was used to separate the desired amount of ^{60}Fe for the half-life measurement. Besides iron, the radiochemical analysis indicated also high amounts of ^{60}Co, ^{44}Ti and its daughter ^{44}Sc, as well as ^{55}Fe in the copper material. More information on the nuclide inventory and the chemical separation is given by Schumann and Neuhausen [19]. Since the measurement of the ^{60}Fe activity is based on the observation of the grow-in of the activity of its daughter nuclide ^{60}Co, a complete separation of cobalt (about 50 MBq ^{60}Co) in the samples was mandatory. Moreover, ^{44}Ti had to be separated as well, because its daughter isotope ^{44}Sc has a γ-line at 1.16 MeV, which is close to the γ-line of ^{60}Co at 1.17 MeV. Additionally, an efficient separation of nickel had to be achieved, because ^{60}Ni interferes with ^{60}Fe for the MC-ICP-MS measurement. To reach the necessary separation efficiency, dedicated radiochemical procedures were applied. First, the copper chips were dissolved in concentrated nitric acid. As carrier, 5 mg of stable iron and cobalt, respectively, was added to achieve higher chemical

yields for the corresponding elements. After evaporating the solution to dryness, the residue was dissolved in 7 mol/L hydrochloric acid. In this medium, iron forms a negatively charged chloride complex, which can easily be extracted into diethyl ether, whereas cobalt, titanium, and nickel stay in the aqueous phase. The back extraction was carried out with diluted hydrochloric acid (0.1 mol/L). In order to increase the separation factors, the procedure was repeated twice. A final purification was performed by precipitating Fe as hydroxide with diluted ammonia solution, which gives an additional purification from cobalt and nickel [15]. A decontamination factor of better than 10^7 for ^{60}Co was achieved for the final product. The decontamination factor for Ni is unknown due to the lack of in-house analytical equipment such as ICP-OES capable to determine the concentration of stable nuclides in highly radioactive samples. The chemically purified material was then dissolved in diluted hydrochloric acid and transferred into a glass vial with a diameter of 22 mm (SU860065 with the septum #854996, Sigma-Aldrich Chemie GmbH, Taufkirchen, Germany). This vial (called master sample) was transferred to the Technische Universität München for the determination of the ^{60}Fe activity.

After almost 3.5 years of activity measurement, the master sample returned to PSI for the determination of the number of ^{60}Fe atoms and the total amount of iron. The vial was carefully weighed, the liquid was retrieved from it, and the cleaned and dried vial was weighed again to calculate the total mass of the solution. The septum of the vial was stained with an orange/red substance assumed to be $FeCl_3$. To account for potential iron losses, the activity of the cap was measured. The determined activity was less than one per mille of the master sample activity at that time. During the measurement campaign, a loss of about 0.5 g of the initial mass of 5 g of the master sample was observed, most probably due to evaporation losses through the imperfect sealing of the vial with the rubber seal. From the remaining 4.545(8) g of the master sample, two aliquots were taken for the MC-ICP-MS measurements: aliquot A, 100 mg for isotope dilution mass spectrometry (IDMS) and aliquot B, 400 mg for the determination of the isotopic composition by MC-ICP-MS. Aliquot A was directly diluted and spiked with an enriched material, whereas aliquot B was further chemically treated because the first MC-ICP-MS measurement revealed that nickel had not been completely separated and was of nonnatural isotopic composition. Due to the nonnatural Ni, a mathematical correction of the signal on $m/q \simeq 60$ was impossible. To overcome this, in total 20 μg natural nickel carrier was added to aliquot B to dilute the nickel isotopes to natural composition. The iron fraction was separated by precipitation as hydroxide with ammonia solution and keeping the nickel in the solution as described above. The precipitate was dissolved in diluted hydrochloric acid. After six repetitions of the procedure, the solution was pure enough for an accurate MC-ICP-MS isotope ratio measurement including a nickel correction.

In general, an almost perfect separation of the analyte from potential interfering elements is necessary. The fact that the base materials were exposed to high-energy particle beams or neutron fields results in nonnatural isotopic composition of the matrix. Further by-products from the irradiation or the decay will hamper the accurate determination of the isotopic composition.

Activity measurement

The total activity of ^{60}Fe was determined by measuring time series of γ-count rates representing the grow-in of its daughter isotope ^{60}Co using the two prominent γ-lines at 1.17 and 1.33 MeV. The details of the activity measurements are described in [1].

Number of atoms measurement

Instrumentation

In order to calculate the number of ^{60}Fe atoms, the total iron amount and the isotopic composition of the master sample have to be determined. The method of choice for high-accuracy concentration measurements is IDMS. Thanks to the rather large amount of ^{60}Fe the determination of $N_{^{60}\text{Fe}}$ could be performed by a multiple-collector inductively coupled plasma mass spectrometer (*Neptune* MC-ICP-MS, Thermo Scientific, Bremen, Germany). The instrument is equipped with nine Faraday cups (FCs) and one secondary electron multiplier (SEM) which can be operated instead of the central FC. The system offers nine amplifiers with 10^{11} Ω resistors and one with a 10^{10} Ω resistor. The amplifiers can be connected to any of the FCs via the *virtual amplifier matrix* by the software. This feature was used to extend the dynamic range by deploying the low gain amplifier (10^{10} Ω) for the most abundant isotope. Further details of the system are described elsewhere (see, e.g., [20]). For sample introduction, an Apex high-efficiency desolvating system with a PFA-ST micro-flow nebulizer operated in free aspiration mode with an uptake of 65 μL/min was used (both Elemental Scientific, Omaha, NE, USA). The transport efficiency is enhanced up to one order of magnitude by this introduction system. To further homogenize the aerosol, the outlet of the Apex was connected to a stable sample introduction spray chamber (SSI, Thermo Scientific, Bremen, Germany). As torch assembly, a Fassel-type torch with a narrow bore sapphire injector (AHF, Tübingen, Germany) was used. In order to operate the nebulizer at constant conditions, an additional gas flow was introduced between the SSI chamber and the injector by means of a laminar flow

additional gas adapter (AHF, Tübingen, Germany). The interface was equipped with custom-built aluminum cones instead of standard nickel cones to avoid elevated background for nickel. The detailed operating and acquisition parameters are given in Table 1.

The gain factors of the amplifiers were calibrated on a daily basis. In order to ensure that all detector–amplifier chains provide identical data, the inter FC efficiency deviation was determined in earlier experiments. In this experiment, one isotope pair is measured on several detector pairs. Assuming that all detector–amplifier chains have identical efficiencies, the results should be identical within the theoretical precision of the instrument, which is better than 10 ppm. This was the case in all experiments carried out. In the same experiments, the amplifier decay parameters were updated. These parameters account for variations in the signal decay of a particular amplifier due to differences in the discharge characteristics of the FC through the feedback resistor. The yield of the SEM was determined prior to its use; the retarding potential quadrupole lens—to reduce abundance sensitivity—was not employed in these experiments. The SEM dead time was determined to be 24 ns as described in detail by Richter et al. [21].

Materials and methods

The hydrochloric and nitric acid used throughout the experiment were of semiconductor VLSI grade (Sigma-Aldrich, Buchs, Switzerland). No further purification of the acids was performed. High-purity water was obtained from a Milli-Q ultrapure water purification system (Millipore Corporation, Billerica, MA, USA). As certified reference material (CRM) for the calibration of the mass spectrometer, a pure iron (IRMM-014) from the Institute for Reference Materials and Measurements, Geel, Belgium was applied [22]. A certified ^{57}Fe-enriched material [23] served as spike for isotope dilution. Both materials were dissolved in PTFE beakers with concentrated hydrochloric acid; the isotopic composition of either material is provided in Table 2. After dissolution, the material was quantitatively transferred to high-purity PFA bottles and the concentration was adjusted by gravimetric means to approximately 1,000 mg/L using a lab scale (Mettler AT261, Mettler Toledo, Greifensee, Switzerland). For the MC-ICP-MS measurement, the sample has to be transferred to a defined matrix to avoid possible biases between sample and reference materials. Therefore, the purified ^{60}Fe solution of the master sample was diluted to approximately 4 μg/g with 3 % (*w/w*) nitric acid to achieve ≃50 V on the major isotope for the measurement. The certified reference material was prepared in the same manner to match the matrix and achieve similar signal intensities. The nitric acid used for all dilutions was freshly prepared on a daily basis and also analyzed as blank. As wash solution between runs, first 5 % (*w/w*) hydrochloric acid for two minutes followed by 5 % (*w/w*) nitric acid were used until a background signal of less than 1 mV for ^{56}Fe was obtained.

From aliquot A, five solutions were prepared for IDMS with different spiking ratios $^{57/56}$Fe = 0.75–1.5 to eliminate systematic errors. The concentration of a sample c_x with the sample mass m_x and the mass of added spike m_y as well as their respective relative molecular weights $M_{x,y}$

Table 1 Operating and acquisition parameters of the *Neptune* MC-ICP-MS

Operation	
Forward power	1,350 W
Guard electrode	Grounded
Gas flows:	
Cool	14.6 L min^{-1}
Auxiliary	0.9 L min^{-1}
Sample	0.8 L min^{-1}
Make-up	0.3–0.5 L min^{-1}
Injector	Sapphire (narrow bore)
Spray chamber	APEX HF
Cones:[a]	
Sampler	Aluminum 0.9 mm
Skimmer	Aluminum 1.0 mm
Data acquisition	
Collection mode	Static for CRM and IDMS, dynamic for sample
Amplifier resistor	10^{11} and 10^{10} Ω (L1)
Amplifier rotation	Off
Resolution	≃2,400, medium
Acquisition time	4.194 s
Cycles/block	7
Blocks	21
Blank	5 blocks (on peak)
Background	Defocus

[a] Custom design by PSI

Table 2 Isotopic composition of the certified reference materials

Isotope	Abundance in % (*n/n*)	
	IRMM-014	^{57}Fe enriched material
^{54}Fe	5.845(23)	0
^{56}Fe	91.754(24)	3.11
^{57}Fe	2.1192(65)	95.10
^{58}Fe	0.2818(27)	1.79

can be calculated according to Eq. 2, with the measured isotope ratios $R_{x,y,b}$ of the sample (x), spike (y), and blend (b), respectively, and the sum of all isotope ratios $\sum_i R_{ix}$ of the sample (x) and spike (y), respectively.

$$c_x = c_y \cdot \frac{m_y}{m_x} \cdot \frac{M_x}{M_y} \cdot \frac{R_y - R_b}{R_b - R_x} \cdot \frac{\sum_i R_{ix}}{\sum_i R_{iy}} \tag{2}$$

As an analysis procedure, the standard-sample-bracketing method was used, meaning that prior and after each sample, a reference material was measured to account for drift in the mass discrimination over time. All data were corrected for background, mass bias, and isobaric interferences. For mass bias correction, the exponential law (see Eq. 3) was used, which was found to fit the actual response of the *Neptune* MC-ICP-MS in the mass region of Fe best.

$$R_{obs} = R_{cert} \cdot e^{B \cdot \Delta m} \tag{3}$$

Here, R_{obs} denotes the observed ratio, R_{cert} the ratio of the certified reference material, B the mass discrimination factor, and Δm the mass difference of the isotopes.

The data acquisition was carried out either in a dynamic procedure using FCs and the SEM for the sample or in a static procedure for the measurements of the certified reference material and the IDMS measurements; the cup configuration for either acquisition mode is given in Table 3. In the dynamic procedure, the electric and magnetic field of the mass spectrometer is altered to guide ^{57}Fe$^+$, ^{61}Ni$^+$, or ^{62}Ni$^+$ ions to the central detector; the detectors itself are not moving during the procedure. Further, the central detector is switched from FC to SEM. For the static acquisition, ^{57}Fe$^+$ was guided on the central FC. The dynamic approach was necessary due to the very low nickel content which made it essential to apply the SEM instead of a FC to precisely determine the content of ^{61}Ni and ^{62}Ni. These values were used to correct for the contribution of ^{60}Ni to the signal at $m/q \simeq 60$.

Interferences

Due to the violent environment in the plasma, various undesired species are formed in an ICP. Those species are, e.g., polyatomic interferences like ^{40}Ar^{16}O$^+$ or doubly charged ions like Cd^{2+}. Besides these interferences, isobaric overlaps hamper the precise measurement of isotope ratios in MC-ICP-MS. A comprehensive list of interferences in the mass-to-charge range of Fe is provided in Table 4. To overcome the polyatomic and doubly charged interferences, an increase in mass resolution separates the ions according to their slight difference in mass-to-charge ratios (e.g., ^{40}Ar^{16}O$^+$, ^{112}Cd^{2+} vs. ^{56}Fe$^+$, see Table 4).

A separation of isobaric interferences is not possible with commercial ICP-MS systems. However, a mathematical correction given in Eqs. 4a–4d can be applied in case of a known isotopic composition of the interference, with the respective net signal of the species on the left of the equation

Table 4 List of mass over charge ratios of iron isotopes of interest and of potential interfering ions from the ICP

Ion	m/q [u/e]	Ion	m/q [u/e]
^{54}Fe$^+$	53.93906	^{54}Cr$^+$	53.93833
^{53}Cr^{1}H$^+$	53.94793	^{108}Pd^{2+}	53.95140
^{108}Cd^{2+}	53.95154	^{42}Ca^{12}C$^+$	53.95807
^{40}Ar^{14}N$^+$	53.96491	^{40}Ca^{14}N$^+$	53.96512
^{40}K^{14}N$^+$	53.96652		
^{56}Fe$^+$	55.93439	^{55}Mn^{1}H+	55.94532
^{112}Cd^{2+}	55.95083	^{112}Sn^{2+}	55.95186
^{44}Ca^{12}C$^+$	55.95493	^{40}Ar^{16}O$^+$	55.95675
^{40}Ar^{16}O^{1}H$^+$	55.95675	^{20}Ne^{36}Ar$^+$	55.95944
^{42}Ca^{14}N$^+$	55.96114		
^{57}Fe$^+$	56.93485	^{114}Sn^{2+}	56.95084
^{114}Cd^{2+}	56.95113	^{45}Sc^{12}C$^+$	56.95536
^{41}K^{16}O$^+$	56.95619	^{43}Ca^{14}N$^+$	56.96129
^{58}Fe$^+$	57.93273	^{58}Ni$^+$	57.93534
^{116}Sn^{2+}	57.95032	^{116}Cd^{2+}	57.95183
^{46}Ti^{12}C$^+$	57.95208	^{46}Ca^{12}C$^+$	57.95314
^{20}Ne^{38}Ar$^+$	57.95462	^{22}Ne^{36}Ar$^+$	57.95838
^{44}Ca^{14}N$^+$	57.95801		
^{60}Fe$^+$	59.93352	^{60}Ni$^+$	59.93024
^{60}Co$^+$	59.93327	^{59}Co^{1}H$^+$	59.94047
^{48}Ti^{12}C$^+$	59.94740	^{48}Ca^{12}C$^+$	59.95199
^{22}Ne^{38}Ar$^+$	59.95357	^{20}Ne^{40}Ar$^+$	59.95427
^{46}Ti^{14}N$^+$	59.95516	^{46}Ca^{14}N$^+$	59.95622

The masses of the molecule ions were deduced from the recommended atomic masses [24, 25] of the constitute isotopes and the mass loss of electrons according to the indicated ion charge using the recommended electron mass given in [26]

Table 3 Cup configuration for the static and dynamic data acquisition

Amplifier (Ω)	10^{11}	10^{11}	10^{10}	10^{11}	10^{11}	10^{11}	10^{11}
Cup	L3	L2	L1	C	H1	H2	H3
Static/	^{54}Fe	^{55}Fe	^{56}Fe	^{57}Fe	^{58}Fe	^{60}Fe	^{61}Ni
dynamic		^{55}Mn		FC		^{60}Ni	
Dynamic				^{60}Fe	^{61}Ni		
only				^{60}Ni	SEM		
Dynamic			^{60}Fe		^{62}Ni		
only			^{60}Ni		SEM		

derived from the measured intensity Sig(i) and the natural abundance Abu(i).

$$^{60}\mathrm{Ni}_{^{61}\mathrm{Ni}} = \mathrm{Sig}(61) \cdot \mathrm{Abu}(^{60}\mathrm{Ni})/\mathrm{Abu}(^{61}\mathrm{Ni}) \quad (4a)$$

$$^{60}\mathrm{Ni}_{^{62}\mathrm{Ni}} = \mathrm{Sig}(62) \cdot \mathrm{Abu}(^{60}\mathrm{Ni})/\mathrm{Abu}(^{62}\mathrm{Ni}) \quad (4b)$$

$$^{60}\mathrm{Ni} = \left(^{60}\mathrm{Ni}_{^{61}\mathrm{Ni}} + ^{60}\mathrm{Ni}_{^{62}\mathrm{Ni}}\right)\Big/2 \quad (4c)$$

$$^{60}\mathrm{Fe} = \mathrm{Sig}(60) - ^{60}\mathrm{Ni} \quad (4d)$$

Uncertainties

All indicated uncertainties for MC-ICP-MS measurements as well as the weightings in the course of isotope dilution mass spectrometry are expanded uncertainties $U = k \cdot u$ where u is the combined standard uncertainty estimated following the ISO/BIPM Guide to the Expression of Uncertainty in Measurement [27, 28]. They are given in parentheses and include a coverage factor $k = 1$.

Results and discussion

One of the most critical parts in high-accuracy isotope ratio determinations of Fe is the proper accountancy of interferences. In Fig. 1, an example of a mass spectrum acquired with medium resolution ($R = 2,400$) mode is given. The nature of the *Neptune* MC-ICP-MS explains that the peaks are not baseline separated because the spectrometer is not equipped with an exit slit. Therefore, it is referred to as "pseudo high resolution."

Since isobaric interferences require resolutions much higher than any sector field instrument can provide, a mathematical correction has to be performed. An example for mathematically corrected data is given in Fig. 2. Large
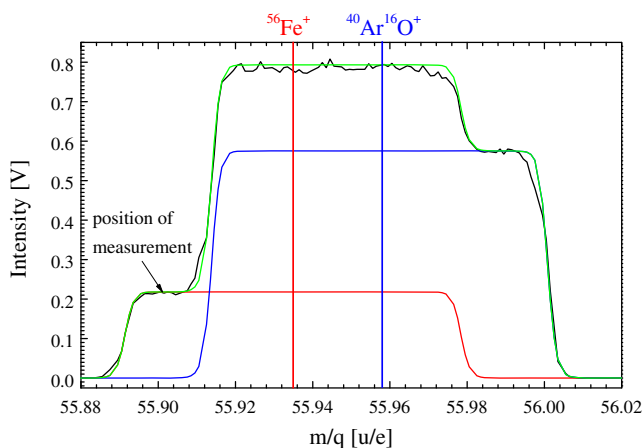

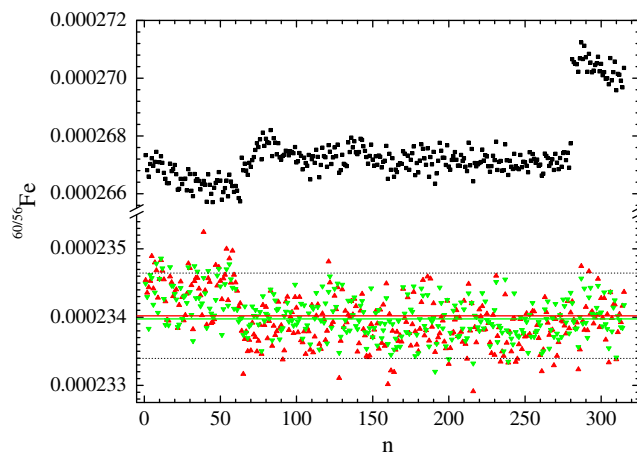**Fig. 2** Data without (*black squares*) and with mathematical nickel correction by ^{61}Ni (*red up triangles*) and ^{62}Ni (*green down triangles*). Each point represents a measurement with an integration time of 4.194 s

efforts were undertaken to dilute the unknown nickel with natural nickel to achieve <0.1 % deviation from natural isotopic composition, monitored by the $^{61/62}$Ni isotope ratio.

The isotopic composition of the master sample derived from the acquired data is listed in Table 5. From this data, the relative molecular weight of the master sample was deduced to be $M_x = 55.9022(33)$ g/mol. For IDMS, an enriched material with a well-known concentration and isotopic composition is mandatory. Unfortunately, the certificate of the enriched ^{57}Fe material did not mention a relative molecular weight. Therefore, this entity was calculated from the given isotopic composition (Table 2) with the assumption of an uncertainty in the last digit to $M_y = 56.922(10)$ g/mol. The same assumption was also made for the calculation of R_y. The concentration of the solution was 0.9472(52) mg Fe/g, based on the assumption of pure enriched iron, a weight of the enriched material of 0.09932(54) g, and a total mass of the prepared solution of 104.85234(54) g. This value was confirmed with a commercial iron reference material from Sigma-Aldrich by reverse IDMS. The confirmation result of 0.9423(43) mg Fe/g is in good agreement with the theoretical value. From this

Fig. 1 Mass scan with medium resolution entrance slit. The *black line* represents the measured intensity, the *red*, *blue*, and *green lines* represent the fitted data for ^{56}Fe$^+$, ^{40}Ar^{16}O$^+$, and the sum of both, respectively

Table 5 Isotopic composition of the master sample

Isotope	Abundance [% (n/n)]	Atomic mass [u] [29]
^{54}Fe	6.033(19)	53.9396147(14)
^{56}Fe	87.4990(50)	55.9349418(15)
^{57}Fe	4.2076(68)	56.9352983(15)
^{58}Fe	2.2397(70)	57.9332801(15)
^{60}Fe	0.02048(12)	59.934072(4) [24, 25]

Table 6 Results of the quantification by IDMS

n	R_b	m_x ($n = 5$)	m_y ($n = 5$)	$N_{Fe} \times 10^{18}$/g	$N_{^{60}Fe} \times 10^{15}$/g
1	0.747264(28)	2.02054(10)	0.57938(12)	6.309(37)	1.292(11)
2	0.958649(42)	2.01720(9)	0.75904(37)	6.307(37)	1.292(11)
3	1.250172(25)	2.02190(9)	1.01413(16)	6.308(37)	1.292(11)
4	1.359976(21)	2.01593(16)	1.21651(18)	6.311(37)	1.293(11)
5	1.482613(54)	2.02597(11)	1.11308(13)	6.309(37)	1.292(11)

stock solution, a working solution was prepared by diluting 0.30179(12) g of stock solution to 32.59509(16) g with 3 % (w/w) nitric acid. Due to the high concentration of the master sample, a dilution was performed before the five blend solutions for IDMS were prepared. For this purpose, aliquot A (0.09949(21) g) was added to a 3 % (w/w) nitric acid; the total mass of the solution was 14.99721(21) g. This solution was then used for each of the five blend solutions; the weighing results of the spiking are given in Table 6. The total number of ^{60}Fe atoms $N_{^{60}Fe} = 5.873(50) \times 10^{15}$ was derived from the total number of iron atoms N_{Fe}.

The half-life was calculated using Eq. 1. From the total activity of the master sample $A_{^{60}Fe} = 49.57(53)$ Bq [1] and the total number of ^{60}Fe atoms $N_{^{60}Fe} = 5.873(50) \times 10^{15}$ atoms, a half-life of ^{60}Fe of $(2.62 \pm 0.04) \times 10^6$ a was deduced. The reported half-life has higher precision than the value reported in [12] ($1.49(27) \times 10^6$ a) and differs considerably. Improvements of the present measurement are due to the larger activity, the long cooling time of the material, as well as the development of more sophisticated measurement techniques. The major type-B uncertainty stems from the activity determination, with contributes about 95 %. The rest of the type-B uncertainty is shared by the weighing and the certified reference materials for the MC-ICP-MS. Almost two-thirds of the type-A uncertainty are contributed by the MC-ICP-MS measurements, the rest is based on the fitting uncertainty of the activity measurement.

Conclusion

The numerous applications of advanced mass spectrometric techniques for half-life determinations in the recent past are remarkable, with the determination of the ^{60}Fe half-life being an excellent example for the high potential of MC-ICP-MS in this research field. For certain applications, the ability to produce highly accurate isotopic and quantitative information makes MC-ICP-MS superior to the well-established accelerator mass spectrometry in half-life determinations. The key to utilizing MC-ICP-MS for the task is the availability of exotic radionuclides in large quantities, which was one of the limiting factors in the past. Due to the combination of this with the recent development of even more sensitive MC-ICP-MS system, more remeasurements of half-lives can be expected in the future.

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