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Corresponding Author : Karen Ostdiek

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Towards a Measurement of the Half-Life of ^{60}Fe for Stellar and Early Solar System Models

K. Ostdiek¹, T. Anderson¹, W. Bauder^{1,2}, M. Bowers¹, P. Collon¹, R. Dressler³, J. Greene², W. Kutschera⁴, W. Lu¹, M. Paul⁵, D. Robertson¹, D. Schumann³, M. Skulski¹, A. Wallner⁶

¹ University of Notre Dame, Notre Dame, Indiana, 46556 USA.

² Argonne National Laboratory, 9700 South Cass Avenue, Lemont, Illinois, 60439, USA.

³ Paul Scherrer Institute - Laboratory for Radiochemistry and Environmental Chemistry, 5232 Villigen, Switzerland.

⁴ Vienna Environmental Research Accelerator Laboratory, Waehringer Strasse 17, 1090 Vienna, Austria.

⁵ Racah Institute of Physics, Hebrew University, Jerusalem, Israel, 91904.

⁶ The Australian National University, Canberra, ACT 0200, Australia.

Radioisotopes, produced in stars and ejected into the Interstellar Medium, are important for constraining stellar and early Solar System (ESS) models. In particular, the half-life of the radioisotope, ^{60}Fe , can have an impact on calculations for the timing for ESS events, the distance to nearby Supernovae, and the brightness of individual, non-steady-state ^{60}Fe gamma ray sources in the Galaxy. A half-life measurement has been undertaken at the University of Notre Dame and measurements of the $^{60}\text{Fe}/^{56}\text{Fe}$ concentration of our samples using Accelerator Mass Spectrometry has begun. This result will be coupled with an activity measurement of the isomeric decay in ^{60}Co , which is the decay product of ^{60}Fe . Preliminary half-life estimates of $(2.53 \pm 0.24) \times 10^6$ years seem to confirm the recent measurement by Rugel, et al. [1].

1. INTRODUCTION

The astrophysical *s*-process (slow neutron capture process) is responsible for producing roughly half of the isotopes above iron. One such isotope is ^{60}Fe , produced by double neutron capture on stable ^{58}Fe . The two most likely sites for this process are massive Asymptotic Giant Branch (AGB) stars and Core Collapse Supernovae (CCSNe), [2]. Both AGB and CCSNe stars lose their outer layers in late stages of star evolution through strong stellar winds and explosions respectively. Each process will eject radioactive isotopes, such as ^{60}Fe , into the Interstellar Medium (ISM). This ejected material can travel as far as 20 - 100 parsecs in the ISM, depending on factors such as the size of the explosion, the ISM, and the amount ejected, amongst others.

Various observations of the ejected material can point to the ongoing nucleosynthesis in the Galaxy. First of all, observations of gamma ray line emission from radioactive isotopes in the Galaxy, including ^{60}Fe , provide constraints to the models describing the nucleosynthesis in massive stars and SNe and show that these processes are continuous [3]. Secondly, meteorites that were created within 1 million years of the Solar System formation, show lower concentrations of $^{60}\text{Ni}/^{58}\text{Ni}$ than Earth, Mars, and chondrite samples [4]. Enhanced amounts of ^{60}Ni , the stable granddaughter product of ^{60}Fe , on Earth could imply that ^{60}Fe was injected into the protoplanetary disk after the Solar System formation. The timing of the ^{60}Fe Solar System injection and its abundance and distribution would help to test and constrain current solar system models. Thirdly and in more recent history, a nearby SN may have overwhelmed the solar winds and deposited matter on Earth. As shown in Figure 1, a spike in the concentration of $^{60}\text{Fe}/\text{Fe}$ has been observed in ocean crust material that has been independently dated to approximately 3 million years ago using the ^{10}Be dating method [5]. This spike in the ^{60}Fe concentration could be evidence of a relatively nearby SN that injected material into the Solar System and can serve as a time indicator for the SN and a test for stellar production models.

The above examples depend on an accurate measurement of the half-life of ^{60}Fe to determine abundances and time scales. However previous measurements of the half-life of ^{60}Fe show a large disagreement. The first experiment published by Roy and Kohman in 1957 [6] measured a half-life of $\sim 3 \times 10^5$ years with a factor of 3 uncertainty, using the isomeric decay in ^{60}Co . The more recent two, published by Kutschera et al. in 1984 [7] and by Rugel et al. in 2009 [1] measured values of $(1.49 \pm 0.27) \times 10^6$ years and $(2.62 \pm 0.04) \times 10^6$ years respectively. Both used the gamma ray transitions in the excited states of ^{60}Ni for the activity measurement. Kutschera et al. used the Accelerator Mass Spectrometry (AMS) technique to measure the number of ^{60}Fe atoms in their sample. Conversely Rugel et al. used Multicollector Inductively Coupled Plasma Mass Spectrometry instead.

2. EXPERIMENTAL METHODS

For half-lives on the order of millions of years, direct measurement of the decay time behavior is not feasible. Instead the measurement relies on determining the number of radioactive atoms of interest in a specific sample and measuring its radioactivity. Then using the following equations, one can deduce the half-life:

$$A = dN/dt = -\lambda \times N = -\frac{\ln(2)}{t_{1/2}} \times N$$

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

where $A = dN/dt$ is the activity or decays per time, N is the number of atoms of interest, and λ is the decay constant related to $t_{1/2}$, the half-life. For the measurement at the University of Notre Dame, the isomeric decay of ^{60}Co will be used for the activity measurement as discussed below, and Accelerator Mass Spectrometry to measure the number of ^{60}Fe atoms, discussed in Section 2.4.

2.1 Decay Scheme

A simplified decay scheme for ^{60}Fe is shown in Figure 2. ^{60}Fe first decays to the 2+ isomeric state in ^{60}Co through β^- decay. This state will decay with a half-life of 10.467 minutes [8]. A small percentage (0.25%) will β^- decay from this excited 2+ state to two of the excited states in ^{60}Ni but most (99.75%) will decay to the ground state in ^{60}Co . This decay will be either by internal conversion (97.9%) or by the emission of a gamma ray of energy 58.6 keV (2.10%) [8,9]. Then the ground state in ^{60}Co will decay to the first 2+ or the 4+ excited states in ^{60}Ni with a half-life of 1925.28 days [8]. Amongst the transitions of the excited state of ^{60}Ni , the most probable are the 1173 keV (4+ to 2+) and the 1332 keV (2+ to 0+ ground state) gamma rays. Therefore, there are two ways to measure the activity of a sample of ^{60}Fe . One way is to measure the decay of the isomeric decay in ^{60}Co . The other way is to measure the transitions in the excited states of ^{60}Ni .

2.2 Samples

The samples used at Notre Dame originated from the Paul Scherrer Institute (PSI) copper beam stop which was bombarded by high energy protons for 12 years, between 1980 and 1992 [10]. The iron material of our experiment had been used as a target to measure the stellar $^{60}\text{Fe}(n, \text{gamma})^{61}\text{Fe}$ cross section [11]. This material was recovered from the target at PSI and split into two batches, one of which was transferred to the Vienna Environmental Research Accelerator Laboratory (VERA) in Vienna, Austria. The other batch remained at PSI for a complementary ^{60}Fe half-life measurement. The Vienna batch contained approximately 3.5×10^{15} ^{60}Fe atoms and was split into two fractions. The larger amount, about 65% of the batch, was used for an activity measurement at the Atominstitut of the Vienna University of Technology. The smaller amount, about 30% of the batch, was used to prepare a dilution sequence with a certificated natural iron solution. Four consecutive dilutions were made, each with a factor of ten lower concentration of ^{60}Fe as the previous one [12]. Each dilution was prepared as a 100 mL solution

with hydrochloric acid. The dilution labels and originally estimated concentrations are shown in Table I. Small amounts of each of these samples were received in powder form to use for the Notre Dame Accelerator Mass Spectrometry experiment and the remaining 13% of the Fe-1 + Hydrochloric (HCl) solution to use for the activity measurement.

2.3 Part 1: Gamma Ray Spectroscopy for Sample Activity

As mentioned in Section 2.1, the isomeric decay in ^{60}Co will be used for the activity measurement instead of the transitions in the excited states of ^{60}Ni . This gives a distinct time advantage because of the slow activity build-up of the ^{60}Ni excited states due to the half-life of ^{60}Co .

The first step was to convert our 13 mL sample of Fe-1 + Hydrochloric (HCl) acid solution into a point source by evaporating most of the HCl. This was done at Argonne National Laboratory (ANL) under the direction of John Greene. The last 0.01 mL of the Fe-1 solution was pipetted onto a Mylar foil. The same technique was used to make a 100 nCi ^{241}Am point source for detector efficiency determination. Both the Fe-1 sample and the ^{241}Am source have a dedicated sample holder for reproducible conditions.

A low-background lead castle counting station was built to house two High-Purity Germanium (HPGe) planar detectors, surrounded on all sides by two layers of lead bricks. Each detector has a measured absolute efficiency of 12% at energies near 59 keV using the ^{241}Am point source. This is due to the thin Beryllium windows and close proximity with the sample (less than 2.5 cm from source to Beryllium window). Data has been taken on this sample for several months, interspersed with background and efficiency measurements.

Figure 3 shows a 24 hour run for both the Fe-1 sample (blue) and the background (orange). The x axis is energy in keV and the y axis is counts per channel. The peak at 64 keV in both runs comes from a decay in the uranium series but is well separated from the $^{60\text{m}}\text{Co}$ decay at 58.6 keV. The

shift in the spectra continuum between the background and sample runs is due to the beta decays of ^{60}Fe and ^{60}Co [13] which have end point energies of 178 keV, for ^{60}Fe and up to 1548.88 keV for ^{60}Co [8]. This shift is accounted for in the background subtraction. The uranium peak after background subtraction is consistent with and without the sample, giving confidence in the activity results. For example, in Figure 3, the activity of the uranium peak is $(7.80 \pm 0.47) \times 10^{-3}$ Bq and $(8.29 \pm 0.33) \times 10^{-3}$ Bq for the background run and the sample run, respectively.

2.4 Part 2: ^{60}Fe Accelerator Mass Spectrometry Measurement

For the measurement of the number of ^{60}Fe atoms in the sample, the Accelerator Mass Spectrometry (AMS) beam line at the Nuclear Science Laboratory at Notre Dame was used. A floor plan is shown in Figure 4. The system makes use of a Multi-Cathode Source of Negative Ions via Cesium Sputtering (MC-SNICS) ion source and an 11 MV FN Tandem accelerator. The AMS beam line contains a Wien Filter, a Browne-Buechner spectrograph, and two gas ionization detectors. For the ^{60}Fe measurements, the spectrograph is run in gas-filled mode, using 1 Torr of nitrogen. As the beam passes through a thin Mylar foil and the gas-filled magnetic field, the individual particles will coalesce around a mean charge state, which depends on the particle's atomic number, Z . Therefore, isobars will take different paths through the magnetic field and can be separated in position when reaching the detectors [14]. This process is shown in more detail in Figure 5. The first detector is a position-sensitive Parallel Grid Avalanche Counter (PGAC) and the second is an Ionization Chamber (IC), split into four sections, for energy loss. The detectors and a movable shield are shown in Figure 6.

Figure 7 shows data from a recent ^{60}Fe AMS experiment at Notre Dame using a beam energy of 95.3 MeV, the 9+ charge state, and $^{56}\text{FeO}^-$ ion source currents below 100 nA. In all three figures, the x axis is position in arbitrary units and the y axis is energy loss in arbitrary units in the third section of the IC. Several samples were measured including three of four of the dilution series mentioned in Section 2.2 and several "blank" Fe_2O_3 cathodes. As noted in Figure 7C,

contaminants from other stable isotopes of iron are not fully separated out after the ion source and before the accelerator. A low energy injection upgrade is planned for the facility in order to eliminate these contaminants and improve the background concentrations achievable by the system. The upgrade will include a 90 degree analyzing magnet, compared to the current 60 degrees, and a 45 degree electrostatic analyzer. Some contamination events may also be from scatter off of the aluminum shield before the detectors in the spectrograph.

3. RESULTS AND FUTURE WORK

Figure 7A shows a clear separation between the isobar ^{60}Ni and ^{60}Fe of an enriched sample. In both Figures 7B and 7C, a shield has been moved in front of the detector. This allows us to block most of the ^{60}Ni isobar from reaching the detectors. Figure 7B is of the enriched sample ($\sim 2 \times 10^{-7}$ concentration) and Figure 7C is blank Fe_2O_3 material. Our background concentration in the current conditions is on the order 10^{-12} $^{60}\text{Fe}/^{56}\text{Fe}$. The measured concentrations of the three enriched samples are shown in Table II. However, these values are about 25% lower than the originally estimated values, shown in Table I.

The Fe-1 sample has not yet been measured by Accelerator Mass Spectrometry (AMS) at Notre Dame. This will need to be measured in order to relate it to the Fe-1 evaporation sample used for the activity measurement. Additionally, during the AMS experiment, the shield position blocked part of the ^{60}Fe peak in the samples that were measured. Attempts were made to correct for this in the final concentrations. Therefore the preliminary estimate for the half-life of ^{60}Fe then is based on the measured $^{60\text{m}}\text{Co}$ activity of (1.28 ± 0.12) Bq and the AMS data published by Wallner, et al. [12]. This paper reports a value of $(1.15 \pm 0.05) \times 10^{15}$ ^{60}Fe atoms in the full Fe-1 sample. Accounting for the fraction of this sample being used at Notre Dame, the half-life of ^{60}Fe is $(2.55 \pm 0.26) \times 10^6$ years, which seems to confirm the longer half-life published by Rugel et al. [1].

Since this experiment, other charge states have been attempted, in particular both 10+ and 11+. The 10+ charge state showed the Bragg peak cross-over in the IC but far too many contaminants with an m/q of 5. The 11+ charge state however looked cleaner and very promising. Currently plans are set on an experiment using 11+ during the summer/fallof 2014-15 at Notre Dame in order to remeasure the previous AMS concentrations and to measure the Fe-1 sample. The possibility of using the second carbon stripper foil setup in the FN tandem is also being explored. This second stripper foil would allow an experiment either at higher beam energies for cleaner background or at the same beam energies done previously but with a lower terminal voltage and therefore more stable running conditions. As the results are still preliminary, a future paper is planned to fully report the activity and updated, independent AMS results.

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Table I: Originally Estimated Concentrations for the VERA dilution series [12].

Sample Name	Nominal Concentrations ($^{60}\text{Fe}/^{56}\text{Fe}$)
VERA Fe - 4	$\sim 2 \times 10^{-9}$
VERA Fe - 3	$\sim 2 \times 10^{-8}$
VERA Fe - 2	$\sim 2 \times 10^{-7}$
VERA Fe - 1	$\sim 2 \times 10^{-6}$

Table II: Measured Concentrations for the VERA dilution series from the Accelerator Mass Spectrometry experiment at the University of Notre Dame.

Sample Name	Measured Concentrations ($^{60}\text{Fe}/^{56}\text{Fe}$)
VERA Fe - 4	$(1.49 \pm 0.04) \times 10^{-9}$
VERA Fe - 3	$(1.52 \pm 0.03) \times 10^{-8}$
VERA Fe - 2	$(1.31 \pm \pm 0.03) \times 10^{-7}$
VERA Fe - 1	Not yet measured

Figure Legends

Figure 1: Measurement of the concentration of $^{60}\text{Fe}/\text{Fe}$ versus age of the ocean crust. The age of the samples used was determined independently by ^{10}Be dating methods [5].

Figure 2: Decay scheme of ^{60}Fe . Thick open lines indicate the most probable decays, both β decays and gamma ray decays. Dotted lines indicate other possible decays that occur with a branching ratio of less than 0.25% [8].

Figure 3: 24 hour runs on the Fe - 1 sample and background. X axis is energy in keV and the y axis is counts. The background peak at 64 keV is from the ^{238}U decay chain, (decay of ^{234}Th to ^{234}Pa).

Figure 4: Nuclear Science Laboratory facility at the University of Notre Dame, including MC-SNICS ion source, FN Tandem accelerator, Analyzing Magnet, Wien Filter, and Spectrograph.

Figure 5: Gas-Filled Magnet Mode: A) Isobars will follow the same path through a magnet field in vacuum. B) After passing through a Mylar foil, the beam will separate into different charge states. However isobars will still follow the same paths. C) After passing through a Mylar foil and nitrogen gas, particles will coalesce around a mean charge state, proportional to the numbers of protons [13].

Figure 6: Drawing of the two detectors in the Spectrograph. The first is a position sensitive PGAC and the second is an IC for energy loss split into four sections. A shield can also be moved in front of the detector to block parts of the beam.

Figure 7: AMS data on enriched ^{60}Fe and blank samples. In all, the x axis is position in arbitrary units and the y axis is energy loss in the third section of the IC in arbitrary units. A) An enriched sample of ^{60}Fe ($\sim 2 \times 10^{-7}$ concentration) with full beam on the detector, 5 minutes and 2 nA of $^{56}\text{FeO}^-$ current at ion source. B) An enriched sample of ^{60}Fe with a shield in place to block most of ^{60}Ni isobar, 15 minutes and 3 nA of $^{56}\text{FeO}^-$ current at ion source. C) A background run in the same settings as A and B using cathode of F_2O_3 material, 15 minutes and 42 nA of $^{56}\text{FeO}^-$ current at ion source.

Figure 1

Figure 2

Figure 3

Figure 3 in Black and White

Figure 4

Figure 4 in Black and White

Figure 5

Figure 5 in Black and White

Figure 6

Figure 6 in Black and White

A)

B)

C)

Figure 7

A)

B)

C)

Figure 7 in Black and White